NEUTRON SCATTERING FROM MAGNETIC SYSTEMS

By

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AKHILESH MAHENDRA

to the

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JUNE 76

CERTIFICATE

Certified that the work presented in this thesis has been carried out by Mr Akhilesh Mahendra under my supervision and has not been submitted elsewhere for a degree

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FOREWARD

Mr Akhilesh Mahendra started his Ph D work under my supervision in July 1969 He successfully completed all Departmental and Institute requirements of the program By April 1972 he concluded his research work and completed the first draft of the thesis Then suddenly, after a brief illness, he passed away

I, personally, would never get over the sense of loss I feel He had a quiet excellence, borne out of a harmonious vision of a brilliant intellect and a gentle heart It has been a pleasure to work with such a colleague and privilege to come across such a good human being

I and my colleagues in the Department felt that the fruits of his labour should come out in the openness of active intellectual life, and be recognized

The Senate of the Indian Institute of Technology, Kanpur, in its meeting dated 23 3 73 decided that his thesis should be examined in the usual manner and if the examiners recommend, he should be awarded a posthumous Ph D degree

During the preparation of the first draft, late Mr. Mahendra and I talked of the various possible changes in the draft However, under the present circumstances, I decided to keep it the way he has left it

I am thankful to various friends and colleagues who took interest in our work, especially to Dr. M P Kapoor, Dr K K Sharma, Dr R K Ray, Dr H S Mani, Dr A.S Parashis, Mr Y S Jain and Mr N L Pathak

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SYNOPSIS

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NEUTRON SCATTERING FROM MAGNETIC SYSTEMS

In the present work the effect of the orbital magnetic moment of the ions on the scattering of neutrons from a magnetic solid is studied Both elastic and inelastic cases are considered For the elastic scattering, the analysis of suitable systems by tensor formalism approach and Halpern-Johnson-Trammell method have been worked out bringing out the strength and weakness of either method Inelastic scattering analysis has been made for rare earth crystals, where orbital magnetic moment are unquenched but spin waves are due to localized electrons and hence easy to handle theoretically Both ferromagnetic and antiferromagnetic cases have been worked out However, since this is the first analytical step in this area, anisotropy effect has been neglected and the study has been restricted to the somewhat idealized cases for the most part

In Chapter I, the tensor formalism developed by Johnston for the elastic scattering of neutrons is extended to several

unfilled shells configurations The case of two shells is investigated completely Some examples of the crystals having this configuration are presented and a simple e ample of U^{4+} ion is worked out Finally, the method for the generalization of the theory to a configuration of N-shells is given

In Chapter II, the spherical magnetic form factors of Co^{++} in CoO and Fe^{++} in FeO are calculated including the effect of the unquenched orbital magnetic moment Expansions relative to the 'spin only' case are found to be 11 % for Co^{++} and 9 % for Fe^{++} For CoO this is a major step in the quantitative explanation of the 15-17 % expansion of the experimental curve compared to the Freeman-Watson 'spin only' curve For FeO no good experimental data are yet available for comparison

In Chapter III, the inelastic scattering cross-section of neutrons for a ferromagnetic rare earth crystal is derived, considering the interaction of neutron with the spin as well as the orbital angular momentum of the ions The case considered is that of an ideal rare earth ferromagnet without anisotropy This would be a basic step towards a general theory of scattering intensities of neutron groups for one magnon process Actually two different methods were improvised, keeping in mind the needs towards generalizations in two directions (i) more realistic ferromagnetic cases, (ii) more complicated magnetic structures, e.g., antiferromagnetics and ferrimagnetics Finally, it is

checked that the cross-section reduces correctly to the 'spin only' case in the absence of ionic orbital angular momenta

In Chapter IV, one of the methods of the Chapter III has been extended to determine the inelastic scattering cross-section of neutrons for the ideal antiferromagnetic case with a perfectly ordered ground state

SECTION - A

ELASTIC SCATTERING OF NEUTRONS

CHAPTER - I

TENSOR FORMALISM CALCULATIONS FOR SEVERAL UNFILLED SHELLS CONFIGURATIONS

1 1 INTRODUCTION

The theory of elastic scattering of neutrons by magnetic salts was developed by Halpern and Johnson (1939) with the assumption that no orbital angular momentum is present in the ion Although the formulae derived by them were used to analyse the neutron scattering data, it was soon realised that orbital angular momentum cannot be ignored for a large number of magnetic salts. Experimental studies by Koehler and Wollan (1953) of some rare earth oxides led Trammell (1953) to calculate the effect of the orbital angular momentum on the neutron scattering cross section Trammell simplified the electron-neutron interaction term to a form which facilitates the calculation of its matrix elements Moreover, using the form of interaction given by Trammell, one can separate the form factor (Blume 1961) in the cross section expression.

However, the Halpern-Johnson-Trammell (hence called HJT) method does not give a general expression for the cross section It is to be calculated separately for each individual ion and, for that matter, for each individual possible wavefunction of the ion in the crystal A realistic crystal wavefunction is complicated due to the presence of a large number of components

which make the calculation of the cross section quite cumbersome The problem is that in the analysis of neutron diffraction data, it is not possible to proceed from the cross section to the wavefunction Usually, cross sections are derived from a large number of possible wavefunctions with a view to match one of them with the experimental data Hence the derivation of information about the ground state wavefunction of the magnetic ion in a crystal by HJT procedure is quite an unwieldy process

Johnston (1956), using the formalism of Racah algebra and irreducible tensor operators, simplified the cross section expressions for a general ground state arising from a single unfilled shell configuration of magnetic ion The expressions obtained by him for the orbital contribution of the cross section contain the same radial integrals as occur in the spin only part of it Further this approach leads to the results which are identical to that obtained by HJT method (Lovesey 1969), but the formalism is more general and better suited for computer calculations

However, Johnston (1966) confined his theory to the single unfilled shell configuration wavefunctions which occur in most of the magnetic ions But there are established evidences of existences of more than one unfilled shell in the ions of some of the salts of the rare earth and actinium group elements (Kuhn 1962) In this chapter, Johnston's approach (1966) is extended to two unfilled shells configuration to facilitate the more realistic

analysis of neutron scattering data and the determination of the ground state of the type of magnetic ions as mentioned in the next section The expressions for the scattering cross section by a single shell are obtained as a particular case of two shells configuration when one of the shells is comoletely filled or completely empty Also, the formulae are simplified using the dipole approximation and the effect of the coupling of the two shells on the scattering is brought out The example of U^{4+} ion is then worked out using the exact formulae and with the same ground state $|L=5, S=1, J=4, M=4\rangle$, arising out of two different configurations, the one-shell $5f^2$ and the two-shell $5f^1\ 6d^1$ This demonstrates clearly the difference between the two-shell and one-shell scattering. Finally, this theory is generalized to N-shells configuration

1 2 EVIDENCE FOR TWO SHELLS CONFIGURATION

We shall present here some of the examples of the magnetic salts having two shells configuration for which the present theor is useful $GdCl_{1\ 6}$ was studied by Mee and Corbett (1965) and in order to explain the observed magnetic moment they assigned to Gd ion in it, a mixture of 40 % [Xe] $4f^7 5d^2$ and 60 % [Xe] $4f^7 5d^1$ electronic configurations with one or the other exhibiting a spin only contribution from 5d electrons According to Hund's rule, the ground states of [Xe] $4f^7 5d^1$ and [Xe] $5f^7 5d^2$ are $|L=2, S=4$ $J=2, M=2\rangle$ and $|L=3, S=9/2, J=3/2, M=3/2\rangle$ respectively for the

isolated ion, and $|L=0, S=4, J=4, M=4\rangle$ and $|L=0, S=9/2, J=9/2, M=9/$ when d electrons give spin only contribution The other likely candidates in rare earth group are La and Ce ... divalent ions should probably be $[Xe]\ 4f^n5d^1$ or close to this configuration Conductivity shown by their di-iodides (Jørgensen 1964) gives evidence for the presence of 5d electrons because 4f electrons, being under strong nuclear attraction, cannot contribute In actinium group, the salts of uranium are most extensively studied by the neutron scattering technique and work has been recently in progress for salts of the other elements of this group Magnetic properties of some of the uranium salts indicate the contribution of two unfilled shells (5f and 6d) to ground state of uranium ions A V Pechennikov et al (1968) studied UTe_2 and UTe_3 below 77°K in anti-ferromagnetic phase and concluded that both 5f and 6d electrons contribute to their magnetic properties Picon and Flahaut (1958), assuming the validity of Curie-Weiss law for the susceptibility, concluded that magnetic moments for US_2, which exists in three distinct crystalline forms, and UOS is due to $6d^2$ electrons Recently structural and magnetic properties of uranium sulphides and selenides were studied by Grønvold et. al (1968) The magnetic moment of U^{4+} in US_2 as determined by them is intermediate between the values expected for $6d^2$ and $5f^2$ configurations assuming spin only magnetism for the former and a very large spin orbit coupling compared to the ligand field effects for the latter Thus it is likely that

magnetism of U^{4+} in these salts may arise from $5f^1 6d^1$ electrons with considerable ligand field effects A scattering calculation with the ground state $|L=5, S=1, J=4, M=4\rangle$ of $5f^1 6d^1$ configuration obtained by Hund's rule is then a first step towards a realistic analysis of the problem For conclusive determination of the ground state of U^{4+} in these salts, accurate neutron scattering experimental data are necessary

Little is known about the low temperature electronic configuration of transuranium ions Recently there have been some attempts to determine the magnetic properties of these salts by neutron diffraction study (Green et al 1970, Cox and Frazer 1967) It is very likely that for some of these ions of actinium series both the 5f and 6d shells contribute to the ground state wavefunction

1 3 THEORY FOR TWO SHELLS CONFIGURATION

Let the configuration consist of $n(= n_1 + n_2)$ electrons which are taken to belong to two single atomic shells of orbital angular momenta l_1 and l_2 respectively Thus, the configuration is $(l_1^{n_1}, l_2^{n_2})$ It will be assumed that each electron in a shell has the same radial wavefunction The complete wavefunction for the system is

$$|\Psi\rangle = | l_1^{n_1} v_1 S_1 L_1, \; l_2^{n_2} v_2 S_2 L_2, \; S M_S \; L M_L \rangle$$

where v is the seniority number

To construct it, we first set up the wavefunctions for the individual shells (Judd 1963) as

$$|\Psi_1\rangle = |l_1^{n_1} \nu_1 S_1 M_{S_1} L_1 M_{L_1}\rangle$$

$$= \sum_{\bar{\nu}_1,\bar{S}_1,\bar{L}_1} \sum_{M_{S_1},M_{L_1}} \sum_{m_{s_1},m_{l_1}} (l_1^{n_1-1} \bar{\nu}_1 \bar{S}_1 \bar{L}_1 | \} l_1^{n_1} \nu_1 S_1 L_1\rangle$$

$$x \langle \bar{L}_1 \bar{M}_{L_1} \; l_1 m_{l_1} | L_1 M_{L_1}\rangle \langle \bar{S}_1 \bar{M}_{S_1} \; s m_{s_1} | S_1 M_{S_1}\rangle$$

$$x \; |S_1 M_{S_1} L_1 M_{L_1}\rangle | s m_{s_1} l_1 m_{l_1}\rangle \qquad (1)$$

and a similar expression for $|\Psi_2\rangle$, representing the wavefunctions for 1st and 2nd shell respectively $\bar{\nu}_1,\bar{S}_1,\bar{L}_1,\bar{M}_{S_1},\bar{M}_{L_1}$ denote the quantum numbers for (n_1-1) electrons Other notations are self-explanatory

The antisymmetric product wavefunction (Condon and Shortley 1953) for the two shells can be written as

$$|\Psi_{12}\rangle = |l_1^{n_1} \nu_1 S_1 M_{S_1} L_1 M_{L_1}, \; l_2^{n_2} \nu_2 S_2 \; S_2 L_2 M_{L_2}\rangle$$

$$= \left[\frac{n_1' \; n_2'}{(n_1+n_2)'}\right]^{1/2} x \; \Sigma \; (-1)^q \; Q | l_1^{n_1} \nu_1 S_1 M_{S_1} L_1 M_{L_1}\rangle$$

$$= |l_2^{n_2} \nu_2 S_2 M_{S_2} L_2 M_{L_2}\rangle \qquad (2)$$

where Q exchanges electrons between $|\Psi_1\rangle$ and $|\Psi_2\rangle$, and $(-1)^q$ is the parity of these exchanges (q being the number of

exchanges) The final wavefunction can be obtained from $|\Psi_1\rangle$ and $|\Psi_2\rangle$ by the use of Clebsch Gordon coefficients as follows

$$|\Psi\rangle = |l_1^{n_1} v_1 S_1 L_1,\ l_2^{n_2} v_2 S_2 L_2,\ S M_S L M_L\rangle$$

$$= \sum_{M_{L_1}, M_{L_2}} \sum_{M_{S_1}, M_{S_2}} \langle L_1 M_{L_1}\ L_2 M_{L_2} | L M_L\rangle \langle S_1 M_{S_1}\ S_2 M_{S_2} | S M_S\rangle |\Psi_{12}\rangle \qquad (3)$$

It can be readily transformed to $|vSLJM\rangle$ basis

1 3 1 Matrix Element of Magnetic Interaction Operator

As shown by Johnston and Rimmer (1969) and more explicitly by Lovesey and Rimmer (1969), the magnetic interaction operator can be partitioned into two parts One gives rise to the neutron spin factor and the other to the crystal factor Further the crystal factor includes both nuclear scattering and electron scattering Clearly, all other terms of our calculation will be exactly the same as derived by Johnston and Rimmer (1969) except for the electron scattering part So the problem is reduced to the evaluation of the following matrix element which essentially gives the electron scattering part (Lovesey and Rimmer 1969),

$$\langle n\ v\ S\ L,\ J\ M |\ D_q^{(1)}\ | n\ v'\ S'\ L',\ J'\ M'\rangle \qquad (4)$$

where

$$\bar{D}^{(1)} = -\sum_\nu \left[e^{i\bar{k}\cdot\bar{r}_\nu}\ \hat{k} \times (\bar{s}_\nu \times \hat{k}) - \frac{1}{\hbar|\bar{k}|}\ e^{i\bar{k}\cdot\bar{r}_\nu}\ (\hat{k} \times \bar{p}_\nu) \right] \qquad (5)$$

Summation of ν runs over all electrons of an ion which are active in the magnetic scattering $\bar{k}$ is the scattering vector, $\bar{s}_\nu$, $\bar{r}_\nu$ and $\bar{p}_\nu$ are the spin, position and linear momentum operators for ν-th electron The n electrons operator $D_q^{(1)}$ can be written as sum of two parts corresponding to the two shells,

$$-D_q^{(1)} = F = \sum_{i=1}^{n} f_i(\bar{r}_i) = \sum_{i=1}^{n_1} f_i(\bar{r}_i) + \sum_{i=n_1+1}^{n} f_i(\bar{r}_i) \qquad (6)$$

Further, $f(\bar{r}) = f_{orb}(\bar{r}) + f_{spin}(\bar{r})$,

where, $$f_{orb.}(\bar{r}) = -\frac{e^{i\bar{k}\cdot\bar{r}}}{|\bar{k}|}(\hat{k}\times\bar{\nabla})_q, \qquad (7)$$

$$f_{spin}(\bar{r}) = e^{i\bar{k}.\bar{r}}(\hat{k} \times (\bar{s} \times \hat{k}))_q \qquad (8)$$

(7) and (8) arise due to the interaction of the spin magnetic moment of neutron with the magnetic field produced by the orbital motion of the atomic electrons and their spin magnetic moments respectively (Trammell 1953)

The evaluation of matrix element (4) requires straightforward but cumbersome algebra Hence only the results with a brief outline of the approach are given here Since F is sum of one particle operators, i.e (6) for cross matrix elements of the form $\langle l_1^{n_1} \nu_1 S_1 M_{S_1} L_1 M_{L_1} | f(\bar{r}) | l_2^{n_2} \nu_2 S_2 M_{S_2} L_2 M_{L_2} \rangle$ to occur in $\langle \Psi | F | \Psi' \rangle$, the exchanges Q and Q' in $|\Psi\rangle$ and $|\Psi'\rangle$ must differ ($|\Psi'\rangle$ is obtained from $|\Psi\rangle$ by priming the quantum

numbers) If they differ by one exchange, the above mentioned matrix elements will have the following factor,

$$\langle l_1 m_{l_1}\ sm_{s_1}|f(\bar{r})|l_2 m'_{l_2}\ sm'_{s_2}\rangle \times \langle l_2 m_{l_2}\ sm_{s_2}|l_1 m'_{l_1}\ sm'_{s_1}\rangle \quad (9)$$

For each additional difference of exchange in Q and Q', these matrix elements contain (9) multiplied with a pair of orthogonality integrals of one electron states belonging to different shells But (9) itself vanishes due to the orthogonality of one electron wavefunctions Hence the cross matrix elements of the type $\langle l_1^{n_1}\ v_1 S_1 M_{S_1} L_1 M_{L_1}|f(\bar{r})|l_2^{n_2}\ v_2 S_2 M_{S_2} L_2 M_{L_2}\rangle$ do not exist Now, using (3) and (6) we obtain for $\langle\psi|F|\psi'\rangle$,

$$\sum_{M_{L_1},M'_{L_1}} \sum_{M_{S_1},M'_{S_1}} \sum_{M_{L_2},M_{S_2}} n_1 \langle l_1^{n_1}\ v_1 S_1 M_{S_1}\ L_1 M_{L_1}|f(\bar{r})|l_1^{n_1} v'_1 S'_1 M'_{S_1}\ L'_1 M'_{L_1}\rangle$$

$$\times \langle L_1 M_{L_1}\ L_2 M_{L_2}|LM_L\rangle\langle S_1 M_{S_1}\ S_2 M_{S_2}|SM_S\rangle\langle L'_1 M'_{L_1}\ L_2\ L_{L_2}|L'M'_L\rangle$$

$$\times\langle S'_1 M'_{S_1}\ S_2 M_{S_2}|S'M'_S\rangle\delta_{S_2,S'_2}\ \delta_{L_2,L'_2}\ \delta_{v_2,v'_2} + \overleftarrow{\sum}_{M_{L_2},M'_{L_2}} \overleftarrow{\sum}_{M_{S_2},M'_{S_2}} \sum_{M_{L_1},M_{S_1}}$$

$$n_2\langle l^{n_2}\ v_2 S_2 M_{S_2} L_2 M_{L_2}|\ f(\bar{r})|l_2^{n_2}\ v'_2 S'_2 M'_{S_2}\ L'_2 M'_{L_2}\rangle\langle L_1 M_{L_1}\ L_2 M_{L_2}|LM_L\rangle$$

$$\times\langle S_1 M_{S_1}\ S_2 M_{S_2}|SM_S\rangle\langle L_1 M_{L_1}\ L'_2 M'_{L_2}|L'M'_L\rangle\langle S_1 M_{S_1}\ S'_2 M'_{S_2}|S'M'_S\rangle\delta_{S_1,S'_1}\ \delta_{L_1,L'_1}$$

$$\times\ \delta_{v_1,v'_1} \quad (10)$$

The second term of (10) can be obtained from the 1st by the

interchange of indices 1 and 2, and multiplying with a phase factor

$$(-1)^{L_2+L_2'+ L+L' + S_2 + S_2' + 2S_1+ S' + S} \qquad (11)$$

which arises from the four Clebsch Gordan coefficients To calculate the first term in (10) we substitute the explicit form of $|l_1^{n_1} v_1 S_1 M_{S_1} L_1 M_{L_1}>$ and $|l_1^{n_1} v_1' S_1' M_{S_1}' L_1' M_{L_1}'>$ according to (1)

$$\sum_{M_{L_1},M_{L_1}'} \sum_{M_{S_1},M_{S_1}'} \sum_{M_{L_2},M_{S_2}} \sum_{\bar{M}_{L_1}} \sum_{\bar{M}_{S_1}} \sum_{m_{s_1},m_{s_1}'} \sum_{m_{l_1},m_{l_1}'} \sum_{\bar{\theta}_1} {}^{-1} n_1(\bar{\theta}_1|_{,}\theta_1)$$

$$(\bar{\theta}_1|\,'\,\theta_1')$$

$$x\ <sm_{s_1}\ l_1 m_{l_1}|f(\bar{r})|sm_{s_1}'\ l_1 m_{l_1}'><\bar{L}_1\bar{M}_{L_1}\ l_1 m_{l_1}|L_1 M_{L_1}><\bar{S}\ \bar{M}_{S_1}\ sm_{s_1}|S_1 M_{S_1}>$$

$$x\ <\bar{L}_1\bar{M}_{L_1}\ l_1 m_{l_1}'|L_1'M_{L_1}'><\bar{S}_1\bar{M}_{S_1}\ sm_{s_1}'|S_1'M_{S_1}'><L_1 M_{L_1}\ L_2 M_{L_2}|LM_L>$$

$$x\ <S_1 M_{S_1}\ S_2 M_{S_2}|SM_S><L_1'M_{L_1}'\ L_2 M_{L_2}|L'M_L'><S_1'M_{S_1}'\ S_2 M_{S_2}|S'M_S'>\ \delta_{S_2,S_2'}$$

$$x\ \delta_{L_2,L_2'}\ \ \delta_{v_2,v_2'} \qquad (12)$$

where $\theta \equiv vSL$

In order to obtain the matrix element in the $|n\theta JM>$ basis we multiply (12) with $<SM_S LM_L|JM>$ and $<S'M_S'L'\bar{M}_L'|J'M'>$ and sum over $M_S, M_L, M_S', \bar{M}_L'$ To simplify (12) we shall deal with the orbital and spin part of $f(\bar{r})$ separately

1.3.2 Orbital Part

The terms in which the matrix elements of the orbital part of $f(\bar{r})$ (i e., (7)) occur can be simplified by expanding (7)

in tensor form and using Racah algebra (Edmond 1960) parallel to Lovesey and Rimmer (1969) Thus, we get the following expression for the orbital part of (12) in $|n\theta JM\rangle$ basis,

$$\sum_{K'',K'} A_1^{12}(k'',K') \sum_{Q'',Q'}{}' \sqrt{4\pi}\, Y_{Q''}^{K''}(k)\langle K'Q'J'M'|JM\rangle\langle K''Q''K'Q'|1q\rangle \quad (13)$$

where,

$$A_1^{12}(K'',K') = (2l_1+1)^2 [L_1][L_1'][L][L'][J']\sqrt{2}\,\delta_{S_1,S_1'}\,\delta_{S,S'}$$
$$\times \delta_{S_2,S_2'}\,\delta_{L_2,L_2'}\,\delta_{v_2,v_2'}\,(-1)^{l_1+L_2+S'+J'+K'}$$
$$\times (-1)^{K'+1}(2K'+1)(2K''+1)^{1/2}\begin{pmatrix} 1 & K'' & K' \\ 0 & 0 & 0 \end{pmatrix}\begin{Bmatrix} 1 & 1 & 1 \\ K'' & K' & K' \end{Bmatrix}$$
$$\times A(K',K',l_1)(\langle j_{K'+1}\rangle_{11}+\langle j_{K'-1}\rangle_{11})\begin{Bmatrix} J' & K' & J \\ L & S & L' \end{Bmatrix}$$
$$\times \begin{Bmatrix} L' & L & K' \\ L_1 & L_1' & L_2 \end{Bmatrix} n_1 \sum_{\bar\theta_1} (-1)^{\bar L_1}(\bar\theta_1|\}\,\theta_1)(\bar\theta_1|\}\,\theta_1')\begin{Bmatrix} L_1' & K' & L_1 \\ l_1 & \bar L_1 & l_1 \end{Bmatrix} \quad (14)$$

where,

$$A(K',K',L_1) = (-1)^{l_1-1}\left[\frac{(l_1+1)(2l_1+3)}{2l_1+1}\right]^{1/2}\begin{pmatrix} l_1 & K' & l_1+1 \\ 0 & 0 & 0 \end{pmatrix}$$
$$\times \begin{Bmatrix} l_1 & 1 & l_1+1 \\ K' & l_1 & K' \end{Bmatrix}, \quad (15)$$

$\langle j_{K'+1}\rangle_{11}$ etc. are the radial integrals for the 1st shell, and,

$$[\,L\,] \equiv (2L+1)^{1/2} \quad \text{and so on.}$$

Similar to a single shell case, we get from (14),

$$\frac{A_1^{12}(K'-1,K')}{A_1^{12}(K'+1,K')} = \left[\frac{K'+1}{K'}\right]^{1/2} , \quad \lambda = 1,2 \qquad (16)$$

which is a consequence of the fact that K'' dependence of (14) is the same as for the corresponding term of one shell configuration

1.3.3 Spin Part

The term containing the matrix element of (8) in (12) can be simplified by a procedure similar to that given by Lovesey and Rimmer (1969) We obtain for the spin part of (12),

$$\sum_{K'',K'} B_1^{12}(K'',K') \sum_{Q'',Q'} \sqrt{4\pi}\, Y_{Q''}^{K''}(k) \langle K'Q'J'\bar{M}'|JM\rangle\langle K''Q''K'Q'|1q\rangle \qquad (17)$$

where,

$$B_1^{12}(K'',K') = \left(\frac{2K'+1}{3}\right)^{1/2}\left[\frac{2}{3}\lambda^{K''} C_1^{12}(K'',K') - \sum_{\bar{K}} \lambda^{\bar{K}} (-1)^{K'} C_1^{12}(\bar{K},K')\right.$$

$$\left. \times (2\bar{K}+1)(2K''+1)^{1/2} (10/3)^{1/2} \begin{Bmatrix} K & K'' & 2 \\ 1 & 1 & K' \end{Bmatrix} \begin{pmatrix} K'' & \bar{K} & 2 \\ 0 & 0 & 0 \end{pmatrix}\right] \qquad (18)$$

and,

$$C_1^{12}(\bar{K},K') = [L_1][L_1'][S_1][S_1'][L][L'][S][S'][J'][K']\sqrt{4\pi}$$

$$\times \delta_{S_2,S_2'}\, \delta_{L_2,L_2'}\, \delta_{v_2,v_2'}\, (-1)^{\bar{K}+S'+L'+J-J'+L_2+S_2+l_1-s}$$

$$\times [s||z^1||s][L_1||y^{\bar{K}}||l_1] \begin{Bmatrix} \bar{K} & L & L \\ L_2 & L_1' & L_1 \end{Bmatrix} \begin{Bmatrix} 1 & S & S' \\ S_2 & S_1' & S_1 \end{Bmatrix}$$

$$x \begin{Bmatrix} S & S' & 1 \\ L & L' & \bar{F} \\ J & J' & K' \end{Bmatrix} \langle J\bar{K}\rangle_{11}\, n_1 \sum_{\bar{\theta}_1} (-1)^{-\bar{L}_1-\bar{S}_1} (\bar{\theta}_1|\, \theta_1)(\bar{\theta}_1|\, \theta_1')$$

$$x \begin{Bmatrix} S_1' & 1 & S_1 \\ s & \bar{S}_1 & s \end{Bmatrix} x \begin{Bmatrix} L_1' & \bar{K} & L_1 \\ l_1 & \bar{L}_1 & l_1 \end{Bmatrix} \qquad (19)$$

where,

$$z_q^1 = s_q \quad \text{and} \quad [s||z^1||s] = [s||s||s] = (3/2)^{1/2}$$

and,

$$[\, l_1||y^{\bar{K}}||l_1\,] = (-1)^{l_1} (2l_1+1) \left(\frac{2\bar{k}+1}{4\pi}\right)^{1/2} \begin{pmatrix} l_1 & \bar{K} & l_1 \\ 0 & 0 & 0 \end{pmatrix}$$

Since (18) is similar to that for a single shell configuration, it immediately follows that (Lovesey and Rimmer, 1969),

$$\frac{B_1^{12}(K'-1,\, K')}{B_1^{12}(K'+1,\, K')} = \left(\frac{K'+1}{K'}\right)^{1/2}, \quad 1 = 1,2 \qquad (20)$$

No integrals of the type $\langle J_K\rangle_{12}$ coupling the radial part of wavefunction of different shells occur due to the absence of cross matrix elements of the form $\langle l_1^{n_1} v_1 S_1 M_{S_1} L_1 M_{L_1} | f(\bar{r}) | l_2^{n_2} v_2 S_2 M_{S_2} L_2 M_{L_2}\rangle$ i.e., between the different shells in (10)

From (10), (12), (13) and (17) we obtain the following expression for the matrix element (4),

$$\langle n\theta JM|D_q^{(1)}|n\theta' J'M'\rangle$$

$$= -\sqrt{4\pi} \sum_{K'',K'} \left[A_1^{12}(K'',K') + B_1^{12}(K'',K') + A_2^{12}(K'',K') + B_2^{12}(K'',K') \right]$$

$$\times \sum_{Q'',Q'} Y_{Q''}^{K''}(k) \langle K'Q'J'M'|JM\rangle \langle K''Q''K'Q'|1q\rangle \qquad (21)$$

where $A_2^{12}(K'',K')$ and $B_2^{12}(K'',K')$ are obtained from (14) and (18) by the interchange of indices 1 and 2, and multiplying by the phase factor (11)

1.4 A SINGLE SHELL CONFIGURATION

This configuration (l_1^n) is a special case of two unfilled shells configuration $(l_1^{n_1}, l_2^{n_2})$ for $n_2 = 0$, $n_1 = n$

Thus,

$$L_2 = 0,\ S_2 = 0,\ L = L_1,\ S = S_1,\ L' = L_1',\ S' = S_1',$$

$$J = J_1 \quad \text{and} \quad J' = J_1' \qquad (22)$$

where the index 1 denotes the quantum number of the 1st unfilled shell Also we have the equality (Edmond 1960),

$$\begin{Bmatrix} J_1 & J_2 & J_3 \\ 0 & J_3 & J_2 \end{Bmatrix} = (-1)^{J_1+J_2+J_3} \left[(2J_2+1)(2J_3+1) \right]^{-1/2} \qquad (23)$$

Using (22) and (23) in (21), it can be easily chec'ed that single unfilled shell expressions derived by Johnston (1966) and Johnston and Rimmer (1969) are obtained as a special case of two unfilled shells configuration.

1 5 DIPOLE APPROXIMATION

For many neutron scattering experiments, it is sufficient to retain in (21) the terms corresponding to K' = 1, K'' = 0,2 This is ca lled dipole approximation and is adequate for small values of $|\bar{k}|$ such that $\langle J_0 \rangle$ is greater than $\langle J_2 \rangle$

1 5 1 Dipole Approximation for an Isolated Ion

The initial and final states of the target are $|\lambda\rangle$ and $|\lambda'\rangle$ which are obtained by transforming (3) to $|JM\rangle$ basis Thus,

$$|\lambda\rangle = |l_1^{n_1}\theta_1, l_2^{n_2}\theta_2, \theta JM\rangle, \quad |\lambda'\rangle = |l_1^{n_1}\theta_1, l_2^{n_2}\theta_2, \theta JM'\rangle ,$$

and

$$\sum_{\lambda,\lambda'} p_\lambda = \sum_{M,M'} \frac{1}{2J+1} ,$$

p being the probability of occurrence of state $|\lambda\rangle$

Following the steps of Lovesey and Rimmer (1969) and using (21), it can be easily seen that the required cross section for the unpolarised neutrons is

$$\frac{d\Gamma}{d\Omega} = D'^2 \sum_q \sum_{\lambda,\lambda'} p_\lambda \left|\langle \lambda' | D_q^{(1)} | \lambda \rangle\right|^2$$

$$= D'^2 \sum_{K'',K'} \frac{3}{2K'+1} \left\{ A_1^{12}(K'',K') + B_1^{12}(K'',K') + A_2^{12}(K'',K') + B_2^{12}(K'',K') \right\}^2 \qquad (24)$$

where,

$$D' = \left(\frac{\gamma e^2}{m_e c^2} \right) = -0.54 \times 10^{-12} \text{ cm}^{-1}, \quad (\gamma = -1.91) \qquad (25)$$

Now, retaining terms for $\kappa' = 1$, $K'' = 0,2$ in (24) and using (16) and (20),

$$\frac{d\sigma}{d\Omega} = \frac{3}{2} D'^2 \left\{ A_1^{12}(0,1) + B_1^{12}(0,1) + A_2^{12}(0,1) + D_2^{12}(0,1) \right\}^2 \qquad (26)$$

We evaluate $A_1^{12}(0,1)$ from (14),

$$A_1^{12}(0,1) = -\frac{1}{12} \left(\langle J_o \rangle_{11} + \langle J_2 \rangle_{11} \right) \frac{J(J+1)+L(L+1)-S(S+1)}{\left\{ J(J+1) \right\}^{1/2}} \times \frac{L_1(L_1+1)+L(L+1)-L_2(L_2+1)}{L(L+1)} \qquad (27)$$

$A_2^{12}(0,1)$ can be obtained from (27) by interchanging the index 1 of the first shell and index 2 of the second shell (the phase factor (11) being unity)

We get from (19),

$$C_1^{12}(0,1) = -\frac{1}{4} \frac{S_1(S_1+1)+S(S+1)-S_2(S_2+1)}{S(S+1)} \times \frac{J(J+1)+S(S+1)-L(L+1)}{J(J+1)^{1/2}} \langle J_o \rangle_{11} \qquad (28)$$

$C_2^{12}(0,1)$ is obtained from (28) by the same recipe as given for $A_2^{12}(0,1)$

To evaluate $B_1^{12}(0,1)$, $(1 = 1,2)$ from (18) we neglect the terms containing $\langle J_2 \rangle_{11}$ in it. This leads to 15-20 % error in the coefficients of $\langle J_2 \rangle_{11}$ for U^{4+} ion (Section 1.6), which, together with the fact that $\langle J_2 \rangle$ is small compared to $\langle J_0 \rangle$, justifies the present approximation.

So,

$$B_1^{12}(0,1) \simeq \frac{2}{3} C_1^{12}(0,1), \qquad 1 = 1,2 \tag{29}$$

Now, from (26), (27), (28) and (29), we get,

$$\frac{d\sigma}{d\Omega} = D'^2 \frac{J(J+1)}{6} g^2 F^2(k) \tag{30}$$

where,

$$g = 1 + \frac{J(J+1)+S(S+1)-L(L+1)}{2J(J+1)} \tag{31}$$

and,

$$F(k) = F_1(k) + F_2(k), \tag{32}$$

$$F_1(k) = \langle J_0 \rangle_{11} \frac{\{[L_1]'+[L]'-[L_2]'\} x \{[J]'+[L]'-[S]'\}}{2[L]'\{3[J]'+[S]'-[L]'\}} + \frac{\{[S_1]'+[S]'-[S_2]'\} x \{[J]'+[S]'-[L]'\}}{2[S]'\{3[J]'+[S]'-[L]'\}} + \langle J_2 \rangle_{11} \frac{\{[L_1]'+[L]'-[L_2]'\} x \{[J]'+[L]'-[S]'\}}{2[L]'\{3[J]'+[S]'-[L]'\}} \tag{33}$$

where $[L]' = [L]^2 = 2L + 1$

$F_2(k)$ is obtained from $F_1(k)$ by interchanging the shell indices 1 and 2 In the limit $|\bar{k}| \to 0$, $\langle J_0 \rangle_{ii} \to 1$, $\langle J_2 \rangle_{ii} \to 0$, $i = 1,2$ So $F(k) \to 1$ Thus, although the scattering cross section for two shells can be put in the same form as for a single shell, the form factor has completely changed It is interesting to note that in a special case for which $S_1 = S_2$ and $L_1 = L_2$, (32) reduces to,

$$F(k) = \frac{\langle J_0 \rangle_{11} + \langle J_0 \rangle_{22}}{2} + \frac{\langle J_2 \rangle_{11} + \langle J_2 \rangle_{22}}{2} \times \frac{J(J+1)+L(L+1)-S(S+1)}{3J(J+1)+S(S+1)-L(L+1)} \qquad (34)$$

which is same as for a single shell with its radial integrals replaced by their average value over the two shells

1.5.2 Dipole Approximation for General Case

The results given by (21) are simplified in dipole approximation as follows Substituting the explicit form of the wavefunction obtained by transforming (3) to $|JM\rangle$ basis, we get after Racah algebraic simplification,

$$\langle l_1^{n_1}\, \theta_1,\ l_2^{n_2}\, \theta_2,\ \theta JM | (\bar{L}_1)_q | l_1^{n_1}\, \theta_1,\ l_2^{n_2}\, \theta_2,\ \theta JM' \rangle$$

$$= \left[\frac{1}{4}\left\{\frac{2J+1}{J(J+1)}\right\}^{1/2} \frac{J(J+1)+L(L+1)-S(S+1)}{L(L+1)} \times \left\{ L_1(L_1+1) + L(L+1) - L_2(L_2+1) \right\}\right] \times (-1)^{J-M} \begin{pmatrix} J & 1 & J \\ -M & q & M' \end{pmatrix}, \qquad (35)$$

Hence, by Wigner-Eckart theorem,

$$\left[l_1^{n_1}\ \theta_1,\ l_2^{n_2}\ \theta_2,\ \theta J \| \bar{L}_1 \| l_1^{n_1}\ \theta_1,\ l_2^{n_2}\ \theta_2,\ \theta J \right]$$

$$= \frac{1}{4} \left\{ \frac{2J+1}{J(J+1)} \right\}^{1/2} \quad \frac{J(J+1)+L(L+1)-S(S+1)}{L(L+1)}$$

$$x \left\{ L_1(L_1+1) + L(L+1) - L_2(L_2+1) \right\} \qquad (36)$$

So, from (27),

$$A_i^{12}\ (0,\ 1)$$

$$= -\frac{1}{3}\, x\, \frac{1}{(2J+1)^{1/2}}\ (\langle J_0 \rangle_{11} + \langle J_2 \rangle_{11})$$

$$x \left[l_i^{n_1}\ \theta_1,\ l_2^{n_2}\ \theta_2,\ \theta J \| \bar{L}_1 \| l_1^{n_1}\ \theta_1,\ l_2^{n_2}\ \theta_2,\ \theta J \right],\ i = 1,2 \qquad (37)$$

Similar to the orbital part, we obtain the following for the spin part,

$$\langle l_1^{n_1}\ \theta_1,\ l_2^{n_2}\ \theta_2,\ \theta JM | (\bar{S}_1)_q | l_1^{n_1}\ \theta_1,\ l_2^{n_2}\ \theta_2,\ \theta JM' \rangle$$

$$= \left[\frac{1}{4} \left\{ \frac{2J+1}{J(J+1)} \right\}^{1/2} \frac{J(J+1) + S(S+1) - L(L+1)}{S(S+1)} \right.$$

$$\left. x \left\{ S_1(S_1+1)+S(S+1)-S_2(S_2+1) \right\} \right] x(-1)^{J-M} \begin{pmatrix} J & 1 & J \\ -M & q & M' \end{pmatrix} \qquad (38)$$

By Wigner Eckart theorem,

$$\left[l_1^{n_1}\ \theta_1,\ l_2^{n_2}\ \theta_2,\ \theta J \| \bar{S}_1 \| l_1^{n_1}\ \theta_1,\ l_2^{n_2}\ \theta_2,\ \theta J \right]$$

$$= \frac{1}{4} \left\{ \frac{2J+1}{J(J+1)} \right\}^{1/2} \quad \frac{J(J+1)+S(S+1)-L(L+1)}{S(S+1)}$$

$$x \left\{ S_1(S_1+1) + S(S+1) - S_2(S_2+1) \right\} \qquad (39)$$

and from (28),

$$C_1^{12}(0,1) = -\frac{1}{(2J+1)^{1/2}}\langle J_0\rangle_{11}\left[l_1^{n_1}\theta_1, l_2^{n_2}\theta_2, \theta J||\bar{S}_1||l_1^{n_1}\theta_1, l_2^{n_2}\theta_2\,\theta J\right]$$

$$1 = 1, 2 \qquad (40)$$

From (29),

$$B_1^{12}(0,1) = -\frac{2}{3}\frac{1}{(2J+1)^{1/2}}\langle J_0\rangle_{11} \times \left[l_1^{n_1}\theta_1, l_2^{n_2}\theta_2, \theta J||\bar{S}_1||l_1^{n_1}\theta_1, l_2^{n_2}\theta_2, \theta J\right] \qquad (41)$$

Now, it can be easily shown that,

$$A_1^{12}(0,1)\langle 1\, Q'JM'|JM\rangle = \frac{1}{3}(\langle J_0\rangle_{11} + \langle J_2\rangle_{11}) \times \langle l_1^{n_1}\theta_1, l_2^{n_2}\theta_2, \theta JM|(\bar{L}_1)_{Q'}|\, l_1^{n_1}\theta_1, l_2^{n_2}\theta_2, \theta JM'\rangle \qquad (42)$$

and,

$$B_1^{12}(0,1) \simeq \frac{2}{3}\langle J_0\rangle_{11}\langle l_1^{n_1}\theta_1, l_2^{n_2}\theta_2, \theta JM|(\bar{S}_1)_{Q'}|l_1^{n_1}\theta_1, l_2^{n_2}\theta_2, \theta JM'\rangle \qquad (43)$$

Assuming that Q' = 0 component gives the dominant contribution to $\langle\lambda|D_q^{(1)}|\lambda'\rangle$, we get from (21),

$$\langle\lambda|D_q^{(1)}|\lambda'\rangle \simeq \langle\lambda|D_q^{(1)}|\lambda\rangle = -\frac{1}{3}\sqrt{4\pi}\langle\lambda|\sum_{1=1}^{2}\left[(\langle J_0\rangle_{11} + \langle J_2\rangle_{11})\bar{L}_1 + 2\langle J_0\rangle_{11}\bar{S}_1\right]_Z|\lambda\rangle \times \left\{Y_0^0(k)\langle 0010|\,10\rangle\,\delta_{q0} + \frac{1}{\sqrt{2}}Y_q^2(k)\langle 2q10|\,1q\rangle\right\} \qquad (44)$$

using the explicit values of spherical harmonics and Clebsch Gordan coefficients occurring in (44), we obtain,

$$\sum_q |\langle \lambda | D_q^{(\perp)} | \lambda \rangle|^2 \simeq \left| \frac{1}{2} \sin\theta \langle \lambda | \sum_{l=1}^{2} (\langle J_0 \rangle_{ll} + \langle J_2 \rangle_{ll}) \bar{L}_l + 2\langle J_0 \rangle_{ll} \bar{S}_l \right| \lambda \rangle|^2 \quad (45)$$

θ is the angle between k and the direction of quantization z Thus the cross section for the scattering of unpolarised neutrons, evaluated in the dipole approximation is

$$\frac{d\sigma}{d\Omega} = \left(\frac{D'}{2}\right)^2 \sum_{\lambda} p_\lambda |\langle \lambda | \sum_{l} \left[(\langle J_0 \rangle_{ll} + \langle J_2 \rangle_{ll}) \bar{L}_l + 2\langle J_0 \rangle_{ll} \bar{S}_l \right]_\perp | \lambda \rangle|^2 \quad (46)$$

where $(. .)$ indicate the component perpendicular to k of the vector operator in the parenthesis The crystal structure factor in (46) is omitted. Thus the interaction operator for the crystal in the dipole approximation can be replaced by

$$\bar{D}^{(\perp)} = \hat{k} \times \bar{D}^{(D)} \times \hat{k} \quad (47)$$

where,

$$\bar{D}^{(D)} = -\frac{1}{2} \sum_{n,d} e^{i\bar{k}\bar{R}_{nd}} \sum_{l=1}^{2} \left[(\langle J_0 \rangle_{ll} + \langle J_2 \rangle_{ll}) \bar{L}_{l,nd} + 2\langle J_0 \rangle_{li} \bar{S}_{l,nd} \right] \quad (48)$$

which is the sum of operators for the two shells $\bar{R}_{nd} = \bar{n} + \bar{d}$ is the position of the nucleus at the d-th site within the unit

cell defined by the lattice vector $\bar{n}$, $\bar{L}_{i,nd}$ and $\bar{S}_{i,nd}$ are total orbital and spin angular momenta of i-th shell for the ion at $\bar{R}_{nd}$

1 5 3 Effect of Coupling of Two Shells on Scattering

When the two shells with ground state $|l_1^{n_1}\ \theta_1\ J_1 M_1\rangle$ and $|l_2^{n_2}\ \theta_2 J_2 M_2\rangle$ are uncoupled, the system is specified with the wavefunction,

$$|\lambda_{12}\rangle = \left[\frac{n_1!\ n_2!}{(n_1+n_2)!}\right]^{1/2} \sum_q (-1)^q\ Q\ |l_1^{n_1}\ \theta_1 J_1 M_1\rangle\ |l_2^{n_2}\ \theta_2 J_2 M_2\rangle \quad (49)$$

which is similar to (2)

Now, it is easily seen that,

$$\langle\lambda_{12}|\ D_q^{(1)}\ |\lambda_{12}\rangle = \langle l_1^{n_1}\ \theta_1 J_1 M_1|\ D_{1,q}^{(1)}\ |\ l_1^{n_1}\ \theta_1 J_1 M_1\rangle + \langle l_2^{n_2}\ \theta_2 J_2 M_2|\ D_{2,q}^{(1)}\ |l_2^{n_2}\ \theta_2 J_2 M_2\rangle \quad (50)$$

where,

$$-D_{1,q}^{(1)} = \sum_{i=1}^{n_1} f_i(\bar{r}_i) \quad \text{and} \quad -D_{2,q}^{(1)} = \sum_{i=n_1+1}^{n} f_i(\bar{r}_i)$$

are the interaction operators for the electrons of 1st and 2nd shells respectively For both the terms in (50), the results of Lovesey and Rimmer (1969) can be used Thus we get in the dipole approximation,

$$\sum_q |\langle\lambda_{12}|D_q^{(1)}|\lambda_{12}\rangle|^2 = \left|\frac{1}{2}\sin\theta \times \sum_{i=1}^{2} \langle\lambda_i|\left\{(\langle J_0\rangle_{ii}+\langle J_2\rangle_{ii})\ \bar{L}_i + 2\langle J_0\rangle_{ii}\ \bar{S}_i\right\}_z|\lambda_i\rangle\right|^2 \quad .(51)$$

where,

$$|\lambda_1\rangle = |l_1^{n_1}\ \theta_1 J_1 M_1\rangle \quad \text{and} \quad |\lambda_2\rangle = |l_2^{n_2}\ \theta_2 J_2 M_2\rangle$$

so the interaction operator is seen to be,

$$\bar{D}(D) = -\frac{1}{2}\sum_{i=1}^{2} g_i F_i(k) \sum_{n,d} e^{i\bar{k}\cdot\bar{R}_{nd}}\ \bar{J}_{i,nd} \tag{52}$$

with,

$$g_i = 1 + \frac{J_i(J_i+1) + S_i(S_i+1) - L_i(L_i+1)}{2J_i(J_i+1)} \tag{53}$$

and,

$$F_i(k) = \langle J_0\rangle_{ii} + \langle J_2\rangle_{ii}\ \frac{J_i(J_i+1)-S_i(S_i+1)+L_i(L_i+1)}{3J_i(J_i+1)+S_i(S_i+1)-L_i(L_i+1)} \tag{54}$$

If the two shells are coupled, it can be shown from (35), (38) and (45) that,

$$\bar{D}(D) = -\frac{g}{2} F(k) \sum_{nd} e^{i\bar{k}\cdot\bar{R}_{nd}}\ \bar{J}_{nd} \tag{55}$$

where g and $F(k)$ are given by (31) and (32) respectively

A comparison of (52) and (55) and eqn (16.26) of Lovesey and Rimmer (1969) reveals that, when the two shells are coupled and the ion is characterised by total angular momentum $\bar{J}$, the scattering cross-section can be put in a form identical to a single shell case The difference occurs in the form factor But for the uncoupled shells the structure of the scattering cross section is quite different from that for a single shell as the scattering

lengths of the two shells are added up

1 6 EXAMPLE OF URANIUM ION, U^{4+}

This example is worked out to consolidate the understanding of the structure and the physical implications of the expressions derived We shall take the neutron polarization $\bar{P}$ parallel to z and $\hat{k}$ perpendicular to z, similar to the case of Tb^{3+} dealt by Lovesey and Rimmer (1969) and Steinsvoll et al (1967) Then we require only the matrix element $\langle \lambda | D_0^{(\perp)} | \lambda \rangle$ for the electron scattering part, $|\lambda\rangle$ being the ground state of the ion Now, taking $|\lambda\rangle = |5f^1 6d^1, L = 5, S = 1, J = 4, M = 4\rangle$ for U^{4+}, we calculated (21) for q = 0 The results are given in the Table 1 It is seen from the intermediate steps in the process of calculation that the ratio of the coefficient of $\langle J_2 \rangle_{11}$ in $B_1^{12}(0, 1)$ to that in $A_1^{12}(0, 1)$ is 13/81 for i = 1 (5f shell) and 13/65 for i = 2 (6d shell)

To illustrate the effect of two shells on scattering as compared to single shell, the same calculation is done with ground state $|5f^2, L = 5, S = 1, J = 4, M = 4\rangle$ The results are listed in Table 2 It is evident that even when the ground terms are identical the cross-section of the two shell case is very different from that of the one shell case for all finite values of $|\bar{k}|$, the coefficients are different, and also extra terms of the kinds $\langle J_1 \rangle_{22} \langle J_{1'} \rangle_{22}$ and $\langle J_1 \rangle_{11} \langle J_{1'} \rangle_{22}$ (1', l = 0,2,4,6) appear However, for $|\bar{k}| \to 0$, $\langle J_0 \rangle_{11} \to 1$ and $\langle J_1 \rangle_{11} \to 0$,

(l = 2,4,6, ll = 11, 22) and the cross-sections tend to be identical The important thing to notice is that as $|\bar{k}|$ slowly increases from zero, the difference in the cross-sections is mainly due to the inequality of the radial integrals of the two shells

To compare the results of the present tensor formalism with those of the HJT method, we recall that the Trammell operator for the scattering of neutrons by electrons is,

$$-\bar{P} \quad \bar{D}^{(\perp)}$$

(apart from a constant factor) where,

$$\bar{D}^{(\perp)} = \bar{D} - \hat{k}\,(\bar{D}.\hat{k}), \tag{56}$$

$$\bar{D} = -\sum_{j} e^{i\bar{k}.\bar{r}_j}\,\bar{s}_j - \frac{1}{4}\left\{h(k\,r_j)\,\bar{l}_j + \bar{l}_j h(\bar{k}\,\bar{r}_j)\right\} \tag{57}$$

and

$$h(\bar{k}.\bar{r}_j) = 2\left\{\frac{d}{dx}\left(\frac{e^x-1}{x}\right)\right\}_{x=i\bar{k}.\bar{r}_j} \tag{58}$$

Clearly, $\bar{D}$ is arbitrary by $\alpha\bar{k}$, where α is a constant But in the present case, i.e., for k z and $\bar{P}$ || z ,

$$\left[\bar{P}.\bar{D}^{(\perp)}\right]_{k\perp\hat{z}} = \left[PD_z\right]\hat{k}_{\perp}\hat{z}$$

where D_z is the z component of (57) without any arbitrariness A calculation is carried out with D_z for $|5f^1\ 6d^1$, L=5, S=1, J=4, M=4>. By expanding this state in terms of one particle wavefunctions and using the method of Condon and Shortley (ibid)

and remembering $\hat{k} \perp \hat{z}$, it can be shown that,

$$\langle \lambda | D_z^{(\perp)} | \lambda \rangle = \langle \lambda | \left[D_q^{(\perp)} \right]_{q=0} | \lambda \rangle$$

$$= - 1\ 03989\ \langle J_0 \rangle_{11} - 0\ 55998\ \langle J_0 \rangle_{22}$$
$$- 1\ 58812\ \langle J_2 \rangle_{11} - 1\ 16974\ \langle J_2 \rangle_{22}$$
$$+ 0\ 28752\ \langle J_4 \rangle_{11} + 0\ 15265\ \langle J_4 \rangle_{22}$$
$$- 0\ 03200\ \langle J_6 \rangle_{11} \qquad (59)$$

The same results are obtained from Table 1 on substituting the values of spherical harmonics for $\hat{k} \perp \hat{z}$ This shows that the two approaches yield identical results for cases with conditions similar to the present one

1 7 N-SHELL CONFIGURATION

The generalization of the treatment for the two shells configuration to N shells configuration is tedious, although straightforward The configuration comprises $n(=n_1 + n_2 + \ldots n_N)$ electrons which are distributed among N shells as $(l_1^{n_1}, l_2^{n_2}, \ldots, l_N^{n_N})$ The wavefunction for the configuration is constructed by the successive coupling of N individual shells wavefunctions each of which is similar to (1), and the procedure analogous to (2) were used to ensure the antisymmetry of the final wavefunction for the exchange of any two electrons We shall denote by $L_{12 \ldots N'}$ and $S_{12 \ldots N'}$ the orbital and spin quantum numbers for the coupled wavefunction for N' shells where the arrangement of suffixes

TABLE 1

Coefficients of $\langle \lambda | D_o^{(1)} | \lambda \rangle$ for $|\lambda\rangle = |5f^1 6d^1, L=5, S=1, J=4, M=4\rangle$

	$\langle J_o \rangle_{11}$	$\langle J_2 \rangle_{11}$	$\langle J_4 \rangle_{11}$	$\langle J_6 \rangle_{11}$	$\langle J_o \rangle_{22}$	$\langle J_2 \rangle_{22}$	$\langle J_4 \rangle_{22}$	$\langle J_6 \rangle_{22}$
$\sqrt{4\pi}Y_o^o$	-0 69328	-1 11395	--	--	-0 37334	-0 77197	--	--
$\sqrt{4\pi}Y_o^2$	0 31003	0 45593	-0.15630	--	0 16695	0 35126	-0 07245	--
$\sqrt{4\pi}Y_o^4$	--	0 03151	0 11964	-0 01541	--	-0 00450	0 05607	--
$\sqrt{4\pi}Y_o^6$	--	--	0 00194	0 01307	--	--	-0 00327	--
$\sqrt{4\pi}Y_o^8$	--	--	--	-0 00022	--	--	--	--

$\langle J_l \rangle_{11}$ and $\langle J_l \rangle_{22}$ ($l = 0,2,4$ and 6) are the radial integrals of 5f and 6d orbitals respectively

TABLE 2

Coefficients of $\langle \lambda | D_o^{(1)} | \lambda \rangle$ for $|\lambda\rangle = |5f^2, L = 5, S = 1, J = 4, M = 4\rangle$

	$\langle J_o \rangle_{11}$	$\langle J_2 \rangle_{11}$	$\langle J_4 \rangle_{11}$	$\langle J_6 \rangle_{11}$
$\sqrt{4\pi} Y_o^o$	-1 06666	-1 75430	---	---
$\sqrt{4\pi} Y_o^2$	0 47708	0.84089	0 07512	---
$\sqrt{4\pi} Y_o^4$	---	-0 04191	-0 04729	0 06226
$\sqrt{4\pi} Y_o^6$	---	---	-0 00833	-0 05427
$\sqrt{4\pi} Y_o^8$	---	---	---	0 00108

represents the order in which shells are combined It should be noted that any other mode of coupling can be obtained from it simply by interchanging the corresponding indices Also, $L_{N'}$ and $S_{N'}$ are the quantum numbers for the N'th shell and no suffix is used for the quamtum numbers of complete wavefunction So the configuration ground state wavefunction is denoted by $|nvSM_S LM_L\rangle$ or $|nvSLJM\rangle$ omitting the intermediate and individual shells quantum numbers to avoid complicated notation

Analogous to (6) we have,

$$F = \sum_{i=1}^{n_1} f_i(\bar{r}_i) + \sum_{i=n_1+1}^{n_1+n_2} f_i(\bar{r}_i) + \quad + \sum_{i=n_1+\cdots+n_{N-1}+1}^{n} f_i(\bar{r}_i) \quad . \quad (60)$$

Now,

$$\langle n\theta JM|F|n\theta' J'M'\rangle$$

$$= \langle n\theta JM|F_{orb.}|n\theta' J'M'\rangle + \langle n\theta JM|F_{spin}|n\theta' J'M'\rangle$$

$$= \sum_{K'',K'} \sum_{i=1}^{N} (A_i^{12\cdots N}(K'',K') + B_i^{12\cdots N}(K'',K')) \sum_{Q'',Q'} \sqrt{4\pi}$$

$$\times Y_{Q''}^{K''}(k) \langle K'Q'J'M'|JM\rangle\langle K''Q''K'Q'|1q\rangle \quad (61)$$

The superscripts 12 N over A and B denote the shells as well as their mode of coupling in the ground state wavefunction Subscript i indicates the shell whose wavefunction occur in the matrix elements of $f(\bar{r})$ in that term These notations are the generalization of that used in (21).

$$A_1^{12\ \ N}(k'',K') = (2l_1+1)^2\,[L][L'][J']\,\sqrt{2}\,\delta_{S,S'}$$

$$\times\,(-1)^{K'+S'+J'+L_1+L'_1+l_1}\;{}_1{}^{K'+1}\;(2K'+1)(2k''+1)^{1/2}$$

$$\times\begin{pmatrix} 1 & K' & K' \\ 0 & 0 & 0 \end{pmatrix}\begin{Bmatrix} 1 & 1 & 1 \\ K'' & k' & K' \end{Bmatrix} A(K',k',l_1)$$

$$\times\,(\langle J_{K'+1}\rangle_{11} + \langle J_{K'-1}\rangle_{11})\begin{Bmatrix} J' & K' & J \\ L & S & L \end{Bmatrix}$$

$$\times\prod_{N'=2}^{N}\Big[(-1)^{L_{12}\,\cdot\,N'-1\;+\;L'_{12}\;\;N'-1\;+\;L_{N'}}\,[L_{12}\;\;N'-1]$$

$$\times\,[L'_{12}\;\;N'-1]\begin{Bmatrix} L'_{12} & N' & L_{12} & N' & K' \\ L_{12} & N'-1 & L'_{12} & N'-1 & L_{N'} \end{Bmatrix}$$

$$\times\,\delta_{S_{12},\;N'-1,\;S'_{12}\;\;N'-1}\;\delta_{S_{N'},S'_N,S'_{N'}}\;\delta_{L_{N'},S'_{N'}}\;\delta_{v_{N'},v'_{N'}}\Big]$$

$$\times\,n_1\sum_{\bar\theta_1}(-1)^{\bar L_1}(\bar\theta_1|\}\,\theta_1)(\bar\theta_1|\}\,\theta'_1)\begin{Bmatrix} L'_1 & K' & L_1 \\ l_1 & \bar L_1 & l_1 \end{Bmatrix} \qquad (62)$$

$B_1^{12\ \ N}(K'',K')$ is given by (18) with C_1^{12} replaced by $C_1^{12\ \ N}$

where,

$$C_1^{12\ \cdot N}(K'',K')$$

$$= [L][L'][S][S'][J'][K']\,\sqrt{4\pi}\,(-1)^{K'-S+L+J-J'+l_1+s}$$

$$\times\,[s||z^1||s]\,[l_1||Y^{\bar K}||l_1]\begin{Bmatrix} S & S' & 1 \\ L & L' & \bar K \\ J & J' & K' \end{Bmatrix}\langle J\bar K\rangle_{11}$$

$$\times\prod_{N'=2}^{N}(-1)^{1+L_{N'}+S_{N'}+S_{12}\,\cdot\,N'+S'_{12}\,\cdot\,N'+L_{12}\,\cdot\,N'+L'_{12}\,\cdot N'}$$

$$x \left[L_{12} \quad N'-1 \right] \left[L'_{12} \quad N'-1 \right] \left[S_{12} \quad N'-1 \right] \left[S'_{12} \quad N'-1 \right]$$

$$x \begin{Bmatrix} \bar{K} & L_{12} & N' & L'_{12} & N' \\ L_{N'} & L'_{12} & N'-1 & L_{12} & N'-1 \end{Bmatrix} \begin{Bmatrix} 1 & S_{12} & N' & S'_{12} & N' \\ S_{N'} & S'_{12} & N'-1 & S_{12} & N'-1 \end{Bmatrix}$$

$$x \; \delta_{S_{N'},S'_{N'}} \; \delta_{L_{N'},L'_{N'}} \; \delta_{v_{N'},v'_{N'}} \Big]$$

$$x \; n_1 \sum_{\bar{\theta}_1} (-1)^{-\bar{L}_1-\bar{S}_1} (\bar{\theta}_1 | \} \theta_1)(\bar{\theta}_1 | \} \theta'_1) \begin{Bmatrix} S'_1 & 1 & S_1 \\ s & S_1 & s \end{Bmatrix} \begin{Bmatrix} L'_1 & \bar{K} & L_1 \\ l_1 & \bar{L}_1 & l_1 \end{Bmatrix}$$

(63)

$A_2^{12.\ N}(K'',K')$ and $B_2^{12\ \ N}(K'',K')$ can be obtained from $A_1^{12\ \ N}(K'',K')$ and $B_1^{12\ \cdot N}(F'',K')$ respectively by interchanging the quantum numbers of 1st shell by that of the 2nd shell and multiplying it with the phase factor given by (11) To obtain $A_{N'}^{12.\ N}(K'',K')$ and $B_{N'}^{12\ \ N}(v'',K')$ for $N' > 2$ we note that wavefunction for all the $(N'-1)$ shells which are coupled together before including the N'th shells can formally be treated as a single shell wavefunction and, thus, the ground state wavefunction for the configuration bears formal similarity with $(N-(N'-1)+1)$ i e., $(N-N'+2)$ shells configuration wavefunction on Again, we can take N'th shell wavefunction for the 1st shell and $(N'-1)$ shells coupled wavefunction in the place of the 2nd shell The only difference is that the former depends on the intermediate

and the individual shell quantum numbers as well Hence $A^{12\quad N}_{N'}(K'',K')$ and $B^{12\quad N}_{N'}(K'',K')$ can be obtained from $A^{12\quad (N-N'+2)}_{1}(K'',K)$ and $B^{12.\quad (N-N'+2)}_{1}(K'',K')$ by the following recipe, (a) replace the quantum numbers of 1st and 2nd shell by that of l'th shell and coupled quantum numbers of (N'-1) shells respectively (b) Multiply with additional Kronecker delta functions which occur due to the orthogonality relation of the coupled wavefunction for (N'-1) shells, this is because the wavefunction depends on the intermediate and individual shell quantum numbers also, unlike the 2nd shell wavefunction

Due to (a) of this recipe the original order of coupling of (N'-1) shells wavefunction to N'th shell is reversed So the corresponding Clebsch Gordan coefficients are changed which is not required However, their value can be restored by multiplying with a phase factor similar to (11)

1.8 CONCLUSIONS

Usually a single atomic shell of the ion is considered for the interpretation of the elastic scattering of neutrons by magnetic salts But it will be more realistic to consider two shells configuration for quite a few magnetic salts For more than two unfilled shells configuration the wavefunction of the system depends in general not only on the individual shells quantum numbers but also on the intermediate quantum numbers that occur

in the process of coupling This complicates the determination of ground state itself and so the interpretation of the neutron scattering data will be more difficult However, the configurations with more than two shells seem to be less important in magnetic salts

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CHAPTER - II

HJT CALCULATIONS FOR Co^{++} AND Fe^{++} IN THEIR MONOXIDES

2 1 INTRODUCTION

Spherical magnetic form factor of a transition metal ion is a measure of the charge density distribution of the unpaired electrons of the unfilled 3d-shell and as such is of great importance in the study of physical properties of these ions Halpern and Johnson[1], in their theory of scattering of neutrons by these ions, derived an expression for the magnetic form factor Weiss and Freeman[2] calculated the effects of the nonspherical charge distribution of the 3d-electrons in various crystalline fields and thus made it possible to isolate the spherically symmetric form factor The assumption in both the cases has been that the orbital magnetic moment is completely quenched by the crystalline field and its associated magnetic moment does not contribute to neutron scattering

However, an examination of the g-factor of the transition metal ions in different salts shows that there may be sizeable residual orbital moments present[3] In Co^{++} and Fe^{++} this would be expected since the orbital degeneracy of the ground state is not completely lifted by the cubic crystalline field[4] In Ni^{++} the degeneracy is completely lifted, but the spin-orbit coupling causes the admixture of a higher state in the ground state, giving rise to an orbital moment

Recently the spherically symmetric form factor of Co^{++} in antiferromagnetic Cobaltous Oxide was determined experimentally by Khan and Erickson[5] They found out that the form factor is expanded by 15-17 % compared to that calculated for the 'spin-only' case following Halpern and Johnson The present work is undertaken to determine the effect of orbital contribution to this expansion Since the case of Fe^{++} is similar, this was also studied though no suitable experimental data are available as yet for comparison

Scattering by orbital moments has been considered previously by Trammell[6] and in an elaboration of his work by Odiot and Saint-James[7] They were concerned with rare earth ions, in which the orbital moments were completely unquenched and the effects of the crystalline fields are secondary The transition metal ion worked out in the framework of Trammell's theory is Ni^{++} in antiferromagnetic NiO This was done by Blume[8], who accounted for 4 % of the 17 % expansion of the experimental curve of Alperin[9] relative to the 'spin-only' case as due to orbital effect The case of NiO is simpler compared to that of CoO because of the non-degeneracy of the ground state and the uniaxiality of the spin structure in the antiferromagnetic state (Li's mode A) In the present work Trammell's[6] expression for the orbital contribution to the neutron-ion interaction is used The ground states of Co^{++} and Fe^{++} in their monoxides

below the Ne'el temperature as determined by Kanamori[4] have also been useful

2 2 SPIN STRUCTURES OF CoO AND FeO

Calculation of theoretical magnetic form factor requires a knowledge of the spin structure in the crystal The magnetic unit cell of CoO and FeO in antiferromagnetic state, i e , below the Ne'el temperature, is eight times the crystallographic unit cell and contains 32 magnetic ions The magnetic structure (spin structure) of these crystals is such that each spin has antiparallel partners at vector distances $(0,0,\frac{1}{2})$, $(0,\frac{1}{2},0)$, $(\frac{1}{2},0,0)$, parallel partners at $(\frac{1}{2},\frac{1}{2},0)$, $(\frac{1}{2},0,\frac{1}{2})$, $(0,\frac{1}{2},\frac{1}{2})$ If we call such an antiferromagnetic set of spins a submotive, the 32 spins in a magnetic unit cell form four submotives. Thus the spin structure can be described fully by giving the orientation of four spins each belonging to a different submotive At present the most accepted model of the spin arrangement of CoO is van Laar multispin model[10,11] as given in Table 1 and Figure 1

Spin structure of FeO is collinear as given by Li's model A[12] (Figure 2) which is characterised by the fact that crystallographically unique (111) planes are ferromagnetic sheets and alternate (111) planes have parallel and antiparallel arrangement of their spins

TABLE 1

van Laar Multispin Model for CoO

Origin of the submotive		Direction	Cosines of spins	
1	0 0 0	-0 325	-0 325	+0 888
2	$\frac{1}{4}$ 0 $\frac{1}{4}$	+0 325	-0 325	-0 888
3	$\frac{1}{4}$ $\frac{3}{4}$ $\frac{1}{4}$	+0 325	+0 325	+0,888
4	0 $\frac{3}{4}$ $\frac{3}{4}$	-0 325	+0 325	-0 888

2.3 THE SCATTERING CROSS-SECTION

The differential cross section for the elastic magnetic scattering of unpolarised neutrons into solid angle $d\Omega$ is[1,8]

$$\frac{d\sigma}{d\Omega} = \left(\frac{r e^2}{mc^2}\right)^2 \sum_{q,q'} p_q \left| \sum_{\vec{n}} \exp(i\vec{K}\,\vec{n}) \langle q' | \vec{T}_{\vec{n}} | q \rangle \right|^2 \qquad (1)$$

where $|q\rangle$ and $|q'\rangle$ are the initial and final states of the crystal assumed to have the same energy, $\vec{K} = \vec{k} - \vec{k}'$ is the difference between the initial and final wave vectors, $\hbar\vec{k}$ and $\vec{k}'$ respectively of neutron and p_q is the probability that the state $|q\rangle$ is occupied, $\vec{n}$ is the lattice vector and $r' = -1\ 91$ is the gyromagnetic ratio of neutron $\vec{T}_{\vec{n}}$ represents the interaction of neutron with the electrons of the ion at site $\vec{n}$, and is given by

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Figure 1

Multispin-axis structure of CoO The + and - signs designate the up and down directions into the paper of the z components of the spins The arrows represent the projection of the spins on the ab plane The layers $z = \frac{1}{2}$ and $z = \frac{3}{4}$ can be constructed by reversing the spin directions in the layers $z = 0$ and $z = \frac{1}{4}$, respectively

Figure 1

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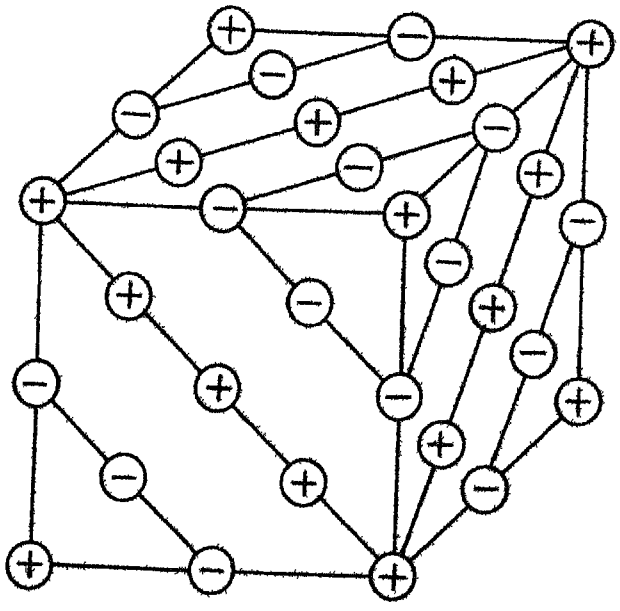
Figure 2

Circles with a + sign represent the Fe^{++} ion with spin up Circles with a - sign represent that with spin down



Li's Model A

PRBLM w /REFNS/FIG. 1 50%

FIGURE 2

$$\vec{T}_{\vec{n}} = \sum_J e^{i\vec{K}\cdot\vec{r}_J}\ \hat{K} \times (\vec{s}_J \times \hat{K}) - \frac{1}{\hbar K} \sum_J e^{i\vec{K}\cdot\vec{r}_J} (\hat{K} \times \vec{p}_J), \qquad (2a)$$

where $\vec{s}_J$ and $\vec{p}_J$ are spin and linear momentum of J^{th} electron, $\vec{r}_J$ is the position of the electron relative to the lattice point $\vec{n}$, $\hat{K}$ is the unit vector in the direction of $\vec{K}$ and the summation is over all electrons of the ion at the lattice site $\vec{n}$ It was shown by Trammell[6] that

$$\vec{T}_{\vec{n}} = \hat{K} \times \vec{Q}_{\vec{n}} \times \hat{K} \qquad (2b)$$

where

$$\vec{Q}_{\vec{n}} = \sum_J \left[e^{i\vec{K}\cdot\vec{r}_J}\ \vec{S}_J + \frac{1}{4} (\vec{I}_J\ f(\vec{K}\cdot\vec{r}_J) + f(\vec{K}\cdot\vec{r}_J)\vec{I}_J \right] \qquad (3)$$

Equation (3) is exact for elastic scattering and holds to a good approximation for inelastic scattering as well,

$$f(\vec{K}\cdot\vec{r}_J) = 2 \left[\frac{d}{dx} \left(\frac{e^x - 1}{x}\right) \right]_{x = i\vec{K}\cdot\vec{r}_J} \qquad (4)$$

$\vec{I}_J$ is the orbital angular momentum of the J^{th} electron of the ion It should be noted that

$$\lim_{\vec{K}\to 0} f(\vec{K}\cdot\vec{r}_J) \to 1 \qquad (5)$$

Clearly, $\vec{Q}_{\vec{n}}$ is arbitrary by $\alpha\hat{K}$, where α is a constant[13] The separation of the scattering cross section into form factor and structure factor is highly dependent on the choice of $\vec{Q}_{\vec{n}}$. The most convenient and physically important choice of $\vec{Q}_{\vec{n}}$ is (3), i.e , for $\alpha = 0$

2 4 DERIVATION OF FORM FACTOR

2 4.1 For Co^{++} in CoO

The derivation of the form factor requires a knowledge of the ground state wavefunction of the magnetic ions in the crystal Kanamori[4] determined the ground state of Co^{++} in CoO He assumed the Li's model A for the spin structure and solved self consistently for the lowest energy state of the Hamiltonian

$$H_o = -2J_1 Z_1 \langle S \rangle S_Z + \lambda' \vec{l} \vec{S} \qquad (6)$$

in triply degenerate ground state under the cubic crystalline field, $\vec{l}$ is the pseudo-angular momentum in the crystalline field and S is the spin of the ground state of Co^{++} Z_1 is the member of next nearest neighbours as nearest neighbours' effect cancels out in the exchange term by virtue of equal number of them being parallel and antiparallel, Z axis is the four fold symmetry axis of the crystal which was also shown to be the spin direction Thus, Kanamori's ground state for Co^{++} is

$$|\Psi_{Co^{++}} = 0\ 875 \left| \tfrac{3}{2}, -1 \right\rangle - 0\ 446 \left| \tfrac{1}{2}, 0 \right\rangle + 0\ 188 \left| -\tfrac{1}{2}, 1 \right\rangle \qquad (7)$$

where the notation $|M_s, M_l\rangle$ is used to specify the wavefunctions on the right hand side ($S=3/2$, $l = 1$ is implied)

But, in view of the fact that van Laar model has replaced the Li's model A for the spin structure of CoO, we should check the validity of (7) We note that in van Laar's model the relative

arrangement of next nearest neighbours is the same as in Li's model A, so the exchange term is unchanged provided the z axis, i.e., the axis of quantization, is taken along the spin direction of the submotive Also $\lambda' \vec{l}\,\vec{S}$ being isotropic[4], the Hamiltonian (6) is unaltered Hence, it follows that (7) can represent the ground state of the Co^{++} ions belonging to any submotive, with the axis of quantization along the spin direction of that ion But (6) is not the complete Hamiltonian, as it neglects the interactions responsible for the relative orientation of submotives Such interaction as calculated by Kanamori for Li's model A is $-Cl_z^2$, ($C \simeq 100\ cm^{-1}$) arising from the magnetostriction The ground state wavefunction (7) changed due to this additional interaction as follows

$$|\Psi'_{Co^{++}}\rangle = 0\ 900\left|\tfrac{3}{2}, -1\right\rangle - 0\ 401\left|\tfrac{1}{2}, 0\right\rangle + 0\ 169\left|-\tfrac{1}{2}, 1\right\rangle \quad (8)$$

The notations are the same as used in (7) It is shown in section 2 4 that the form factor is insensitive to this change in the ground state wavefunction, hence the neglect of $-Cl_z^2$ in (6) is justified as far as the calculation of the form factor is concerned Recently it is indicated by Bertaut[14] from symmetry considerations that, to some extent, Dzialoshinski-Moriya interaction,[15,16] $\underline{D}\,(\vec{S}_1 \times \vec{S}_2)$ is responsible for the orientation of the submotives in van Laar model But this interaction is expected to be only a few per cent of the isotropic exchange

interaction, while it may be effective in relative orientation of the submotive directions, one expects little change in the ionic wavefunctions except, of course, the axis of quantization The calculation of the ground state wavefunctions of Co^{++} in CoO, taking into account all plausible interactions which gave rise to van Laar model is a problem in itself. But, in view of the above arguments it is certain that the ground state of the Hamiltonian (6), i.e , $|\psi_{Co^{++}}\rangle$ is accurate to a good approximation for the calculation of the spherical form factor So we shall use $|\psi_{Co^{++}}\rangle$ in the present work

The pseudo-angular momentum eigen functions in (7) can be transformed to ^4F and ^4P state functions of Co^{++},[17]

$$|\psi_{Co^{++}}\rangle = (1-\beta^2)^{1/2} |^4F\rangle + \beta|^4P\rangle \qquad (9)$$

where

$$\beta = 0\ 185$$

$$|^4F\rangle = 0.875\left(\left[\frac{3}{8}\right]^{1/2}|M_S = 3/2\ ,\ M_L = -1\rangle + \left[\frac{5}{8}\right]^{1/2}|M_S = 3/2,\ M_L = 3\rangle\right)$$

$$- 0.446\ |\ M_S = 1/2\ ,\ M_L = 0\ \rangle$$

$$+ 0\ 188\ \left(\left[\frac{3}{8}\right]^{\frac{1}{2}}|M_S = -\frac{1}{2}\ ,\ M_L = 1\rangle + \left[\frac{5}{8}\right]^{\frac{1}{2}}|M_S = -\frac{1}{2}\ ,\ M_L = -3\rangle\right)$$

and

$$|\,^4P\,> \;=\; 0.875\;|\,M_S = 3/2\,,\; M_L = -1\,> \\ -0.446\;|\,M_S = 1/2\,,\; M_L = 0\,> \\ +0.188\;|\,M_S = -1/2\,,\; M_L = 1\,>$$

The notations used above are the same as in the reference 17 If $|\Psi_{Co^{++}}>$, i e., (7) is in the crystal fixed co-ordinate system whose z axis is along the four fold symmetry axes of CoO, the wavefunction of i^{th} ion is given by,

$$|q_i> \;=\; \mathcal{R}_i\,|\Psi'_{Co^{++}}> \qquad (10)$$

where $\mathcal{R}_i$ is the rotation operator which corresponds to the rotation of the crystal z axis to coincide it with the direction of the magnetic moment of the i^{th} ion

To simplify (1) for the case of Co^{++} in CoO we write, $\vec{r}_j = \vec{a}_j + \vec{r}'_j$, where $\vec{a}_j$ is the position of the nucleus of j^{th} magnetic ion in the magnetic unit cell and $\vec{r}'_j$ is the co-ordinate of j'^{th} electron of jth atom with origin at $\vec{a}_j$ Since we are concerned with the elastic scattering, we have in (1), $|q> = |q'>$, $p_q = 1$, so $\sum_{q,q'} p_q$ drops out We assume that $|q>$, the wavefunction of the magnetic unit cell can be written as a product of state vectors referring to the individual ions (Heitler London approximation), i.e ,

$$|q> \;=\; |q_1>\,|q_2> - - -\,|q_{32}> \qquad (11)$$

Now, it can be readily seen that all ions belonging to the same submotive give rise to the identical contribution which which are simply added up This is due to the fact that negative sign by the reversal of the direction of magnetic moment of ions[8] is counterpoised by another negative sign arising from the exponential factor which depends on the difference of their positions For the orbit l part of the magnetic interaction operator, the same exponential factor as in the spin part has been obtained by going back to (20), which after taking out the exponential factor is converted to (2b) Since each submotive consists of 8 magnetic atoms in a unit cell, (1) can be simplified to yield,

$$\frac{d\sigma}{d\Omega} = \left(\frac{8r_e^2}{mc^2} \right)^2 \sin^2\omega \, | \, \vec{Q} \, | \, \vec{K} = \vec{G} \qquad (12)$$

where

$$\left[\vec{Q}\right]_{\vec{K}=\vec{G}} = \Big[\langle q_1 | \vec{H} | q_1 \rangle + \langle q_2 | \vec{H} | q_2 \rangle \exp\left(2\pi i \, \frac{h+l}{4}\right) + \langle q_3 | \vec{H} | q_3 \rangle \times \exp\left(2\pi i \, \frac{h+3k+2l}{4}\right) + \langle q_4 | \vec{H} | q_4 \rangle \exp\left(2\pi i \, \frac{3k+3l}{4}\right) \Big]_{\vec{K}=\vec{G}} , \qquad (13)$$

$$\vec{H} = \sum_j \left[e^{i\vec{K}\,\vec{r}_j} \, \vec{S}_j + \frac{1}{4} \left(\vec{I}_j f(\vec{K}\,\vec{r}_j) + f(\vec{K}\,\vec{r}_j) \vec{I}_j \right) \right] \qquad (14)$$

J runs over all unpaired electrons of a magnetic ion ω is the angle between $\vec{K}$ and $\vec{Q}$ at $\vec{K} = \vec{G}$, $|q_1\rangle$, $|q_2\rangle$, $|q_3\rangle$ and $|q_4\rangle$ are the ground states of the ions belonging to the four submotives $\vec{G}$ is the reciprocal lattice vector

In the present case it is useful to keep the wave-functions same and transform $\vec{q}$ through similarity transformation by R_i. Therefore,

$$\frac{d\sigma}{d\Omega} = \left(\frac{8re^2}{mc^2}\right)^2 \sin^2\omega \left| \langle \psi_{Co^{++}} | R_1^{-1} \vec{H} R_1 + R_2^{-1} \vec{H} R_2 \exp\left(2\pi i \frac{h+l}{4}\right) + R_3^{-1} \vec{H} R_3 \exp\left(2\pi i \frac{h+3K+2l}{4}\right) + R_4^{-1} \vec{H} R_4 \exp\left(2\pi i \frac{3K+3l}{4}\right) \right| \psi_{Co^{++}} \rangle \Big|^2 \quad (15)$$

Now, making use of the standard identity for the vector operator[18] $\vec{H}$,

$$R^{-1} H_j = \sum_i R^{(3)}_{ji} H_i \quad , \quad (16)$$

where $R^{(3)}$ is the corresponding rotation matrix in the cartesian space, it is shown in the appendix 1 that

$$\langle \psi_{Co^{++}} | R_1^{-1} \left(\sum_j e^{i\vec{K}\vec{r}_j} \vec{S}_j \right) R_1 | \psi_{Co^{++}} \rangle = \hat{K}_1 \langle \psi_{Co^{++}} | \sum_j e^{i\vec{K}\vec{r}_j} S_{j_z} | \psi_{Co^{++}} \rangle \quad (17)$$

And,

$$\langle \psi_{Co^{++}} | R_1^{-1} \left[\frac{1}{4} \Sigma_j (\vec{l}_j f(\vec{K}\vec{r}) + f(\vec{K}\vec{r}) \vec{l}_j) \right] R_1 | \psi_{Co^{++}} \rangle$$
$$= \hat{k}_1 \langle \psi_{Co^{++}} | \frac{1}{4} \sum_j (l_{1_z} f_j + f_j l_{j_z}) | \psi_{Co^{++}} \rangle$$
$$+ \hat{k}'_1 \langle \psi_{Co^{++}} | \frac{1}{4} \sum_j (l_{1_x} f_j + f_j l_{j_x}) | \psi_{Co^{++}} \rangle$$
$$+ \hat{k}''_1 \langle \psi_{Co^{++}} | \frac{1}{4} \sum_j (l_{j_y} f_j + f_j l_{j_y}) | \psi_{Co^{++}} \rangle \quad (18)$$

where $f_j \equiv f(\vec{K}\,\vec{r}_j)$

$\hat{k}_1$, $\hat{k}_1'$ $\hat{k}_1''$ are the unit vectors in the direction of new z,x and y axes respectively

We note the following points in the connection of (18)

(i) $\hat{k}_1'$ and $\hat{k}_1''$ are not unique because there is arbitrariness by a rotation about k_1 which will change $\hat{k}_1'$ and $\hat{k}_1''$

(ii) The 1st term of (18) is predominant for small values of $\vec{K}$ For an extreme case of $\vec{K} = 0$, only the 1st term exists

Since $$\langle \Psi_{Co^{++}} | \frac{1}{4} \sum_j (l_{j_x} f_j + f_j l_{j_x}) | \Psi_{Co^{++}} \rangle \qquad (19)$$

and $$\langle \Psi_{Co^{++}} | \frac{1}{4} \sum_j (l_{j_y} f_j + f_j l_{j_y}) | \Psi_{Co^{++}} \rangle \qquad (20)$$

do not contain spherically symmetric part (Appendix 2), the spherical form factor which we aim at would not have contribution from the second and third terms of (18) So the omission of these terms does not affect the spherical form factor

After dropping out the second and third term of (18), we use it together with (17) to obtain the following expression for (15),

$$\left(\frac{8r_e^2}{mc^2}\right)^2 \sin^2\omega \; |\langle \Psi_{Co^{++}} | H_z | \Psi_{Co^{++}} \rangle|^2 \times |\hat{k}_1 + \hat{k}_2 \exp\left(2\pi i \frac{h+l}{4}\right) + \hat{k}_3 \exp\left(2\pi i \frac{h+3K+2l}{4}\right) + \hat{k}_4 \exp\left(2\pi i \frac{3K+3l}{4}\right) \qquad (21)$$

Thus starting with the fundamentals and taking proper consideration to the orbital contribution we arrived at an expression for

differential cross section similar to (22) quoted below, which was used by van Laar,[11] Khan and Erickson[5] to analyse the experimental results for CoO

$$\frac{d\sigma}{d\Omega} = \left(\frac{8\pi e^2}{mc^2}\right)^2 \sin^2\omega \left(\frac{\mu_{Co^{++}} f_{Co^{++}}}{2}\right)^2 \times \left|\hat{k}_1 + \hat{k}_2 \exp\left(2\pi i \frac{h+l}{4}\right) + \hat{k}_3 \exp\left(2\pi i \frac{h+3K+2l}{4}\right) + \hat{k}_4 \exp\left(2\pi i \frac{3K+3l}{4}\right)\right|^2 \qquad (22)$$

where, $\mu_{Co^{++}}$ and $f_{Co^{++}}$ are the magnetic moment and magnetic form factor of Co^{++} respectively (22) was obtained from the 'spin only' case by treating the spins classically and replacing the spin moment by the total magnetic moment of the ion Comparing (22) with (21) we can identify the spherical form factor as follows

$$\left[\mu^2_{Co^{++}} f^2_{Co^{++}}\right]_{\text{Sph Part}} = 4\left|\langle \psi_{Co^{++}} | H_z | \psi_{Co^{++}} \rangle\right|^2_{\text{Sph Part}} \qquad (23)$$

It should be noted that comparison of (21) with (22) is valid only for the spherical symmetric part of the form factor due to the approximations involved in neglect of (19) and (20) Taking the limits of both sides for $\vec{K} \to 0$, we get,

$$\mu_{Co^{++}} = \langle \psi_{Co^{++}} | \Sigma_1 (2s_{1z} + l_{1z}) | \psi_{Co^{++}} \rangle = 2\langle \psi_{Co^{++}} | H_z | \psi_{Co^{++}} \rangle_{\vec{K}=0} \qquad (24)$$

as $f_{Co^{++}}(0) = 1$ by normalization condition

Hence,

$$\left[f_{Co^{++}}(K) \right]_{Sph} = \left[\frac{\langle \psi_{Co^{++}} | H_Z | \psi_{Co^{++}} \rangle}{\langle \psi_{Co^{++}} | H_Z | \psi_{Co^{++}} \rangle_{\vec{K}=0}} \right]_{Sph} \qquad (25)$$

2 4.2 For Fe^{++} in FeO

The ground state of Fe^{++} is ^{4}

$$| \psi_{Fe^{++}} \rangle = 0\ 909\ |\ 2, -1\rangle - 0\ 395\ |\ 1,0\rangle + 0\ 135\ |\ 0,1\rangle \qquad (26)$$

Only M_S and M_l values are specified for the kets on the right hand side of (26), S = 2, l = 1 is implied As before, the pseudo-angular momentum eigen function in (26) are transformed to ^{5}D state functions of Fe^{++} to yield,

$$\begin{aligned} | \psi_{Fe^{++}} \rangle = & - 0\ 909\ |\ M_S = 2,\ M_L = 1\ \rangle \\ & - \frac{0.395}{\sqrt{2}} \left(|M_S = 1,\ M_L = 2\rangle - |M_S = 1,\ M_L = 2\rangle \right) \\ & + 0\ 135\ |\ M_S = 0,\ M_L = -1\rangle \end{aligned} \qquad (27)$$

The notations are similar to that used for $|\psi_{Co^{++}}\rangle$

Proceeding parallel to Co^{++}, one can easily derive the form factor for Fe^{++} in FeO Due to the collinear spin structure of FeO, the derivation of the form factor is less involved, as simply an additional negative sign occurs with the magnetic interaction operator $\vec{H}$ for the ions with antiparallel spins[8] As a further consequence of collinearity of the spin structure, we do not require to make approximations similar to those involved

in neglecting (19) and (20) Thus we obtain the following expression for the differential cross section

$$\frac{d\sigma}{d\Omega} = \left(\frac{8re^2}{mc^2}\right)^2 \sin^2\omega \left|\langle \psi_{Fe^{++}} |\vec{H}| \psi_{Fe^{++}} \rangle\right|^2 \left|1 - \exp\left(2\pi i \frac{h+l}{4}\right) + \exp\left(2\pi i \frac{h+3K+2l}{4}\right) - \exp\left(2\pi i \frac{3K+3l}{4}\right)\right|^2 \quad (28)$$

Adopting a procedure similar to the one which led to (22) for Co^{++}, we obtain for Fe^{++} the following expression for the differential cross section

$$\frac{d\sigma}{d\Omega} = \left(\frac{8re^2}{mc^2}\right)^2 \sin^2\omega \left(\frac{\mu_{Fe^{++}} f_{Fe^{++}}}{2}\right)^2 x \left|1 - \exp\left(2\pi i \frac{h+l}{4}\right) + \exp\left(2\pi i \frac{h+3K+2l}{4}\right) - \exp\left(2\pi i \frac{3K+3l}{4}\right)\right|^2 \quad (29)$$

Comparing (28) and (29), we obtain

$$\mu_{Fe^{++}} f_{Fe^{++}}(\vec{K}) = 2\left|\langle \psi_{Fe^{++}} |\vec{H}| \psi_{Fe^{++}} \rangle\right| \quad (30)$$

Since $f_{Fe^{++}}(0) = 1$ by normalization, $\mu_{Fe^{++}}$ is given by $2\left|\langle \psi_{Fe^{++}} |\vec{H}| \psi_{Fe^{++}} \rangle\right|_{\vec{K}=0}$ which follows frpm the limit of (30) for $\vec{K} \to 0$

Hence,

$$f_{Fe^{++}}(\vec{K}) = \frac{\left|\langle \psi_{Fe^{++}} |\vec{H}| \psi_{Fe^{++}} \rangle\right|}{\left|\langle \psi_{Fe^{++}} |\vec{H}| \psi_{Fe^{++}} \rangle\right|_{\vec{K}=0}} \quad (31)$$

which is the complete form factor unlike (25)

2.5 CALCULATION OF FORM FACTOR

2 5 1 For Co^{++} in CoO

We substitute in (25) the explicit form of $|\Psi_{Co^{++}}\rangle$ and H from (9) and (14) respectively Since $\vec{H}$ is sum of one electron operators, we must express $|\Psi_{Co^{++}}\rangle$ in terms of one electron wavefunctions There are three holes in the 3d shell of Co^{++}, $|\Psi_{Co^{++}}\rangle$ is expressed in terms of these hole wavefunctions in the appendix 3a Now, (25) is easily calculated to yield the following result,

$$f_{Co^{++}}(\vec{K}) = \frac{1}{1\ 746} \times \Big[0\ 491 \langle 2|e^{i \vec{K} \vec{r}}|2\rangle + 0\ 391 \langle 1|e^{i \vec{K} \vec{r}}|1\rangle + 0\ 348\langle 0|e^{i \vec{K}.\vec{r}}|0\rangle + 0\ 272 \langle 2|f(\vec{K} \vec{r})|2\rangle + + 0.244 \langle 1|f(\vec{K} \vec{r})|1\rangle \Big] \qquad (32)$$

Using the results of appendix 4 for $\langle dm|\ e^{i \vec{K} \vec{r}}|dm\rangle$ and $\langle dm|f(\vec{K} \vec{r})|dm\rangle$, we have for (32),

$$f_{Co^{++}}(\vec{K}) = \frac{1}{1\ 746} \times \Big[1.230 \langle J_0\rangle + 0\ 516 \langle g_0\rangle + \frac{2}{7}(5\pi)^{1/2} Y_{20}(\hat{K}) \cdot (-0\ 105 \langle J_2\rangle + 0\ 300 \langle g_2\rangle) + \frac{2}{7}\sqrt{\pi}\ Y_{40}(\hat{K}) \times (1\ 011 \langle J_4\rangle - 0\ 704 \langle g_4\rangle) \Big] \qquad (33)$$

To separate out the spherically symmetric part of $f_{Co^{++}}(K)$ we retain in (33) only the spherical symmetric part of $Y_{lm}(K)$ given in appendix 5 Thus,

$$f^{s}_{Co^{++}}(K) = \frac{1}{1.746} \times \Big[1\ 230\ \langle J_0 \rangle + 0\ 037\ \langle J_2 \rangle + 0\ 162\ \langle J_4 \rangle + 0\ 516\ \langle g_0 \rangle - 0\ 107\ \langle g_2 \rangle - 0\ 113\ \langle g_4 \rangle \Big] \qquad (34)$$

$$= \frac{1}{1\ 746} \times \Big[1.230\ \langle J_0 \rangle + 0\ 037\ \langle J_2 \rangle + 0.162\ \langle J_4 \rangle + 0.516 \times (\langle g_0 \rangle - \tfrac{1}{2} \langle g_2 \rangle) + 0.151 \times (\langle g_2 \rangle - \tfrac{3}{4} \langle g_4 \rangle) \Big]$$

where

$$f^{s}_{Co^{++}}(K) \equiv \Big[f_{Co^{++}}(K) \Big]_{\text{sph part}} \qquad (35)$$

Now using the relation between $\langle g_L \rangle$ and $\langle J_L \rangle$,[13]

$$\langle g_{L-1} \rangle - \frac{L}{L+1} \langle g_{L+1} \rangle = \frac{2}{L+1} (\langle J_{L-1} \rangle + \langle J_{L+1} \rangle) \qquad (36)$$

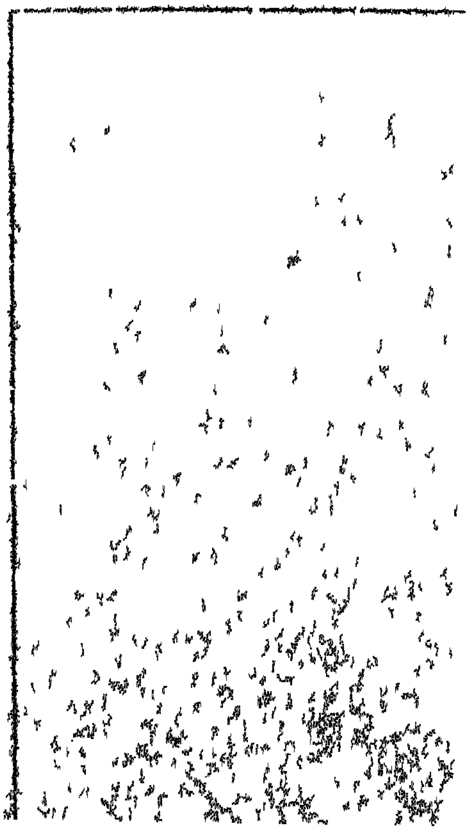
(35) can be written in terms of $\langle J_L \rangle$ only,

$$f^{s}_{Co^{++}}(K) = \langle J_0 \rangle + 0\ 360\ \langle J_2 \rangle + 0.136\ \langle J_4 \rangle \qquad (37)$$

$\langle j_0 \rangle$, $\langle J_2 \rangle$ and $\langle J_4 \rangle$ are tabulated as functions of $\frac{\sin \theta}{\lambda}$ by Watson[19] ($K = \frac{4\pi \sin \theta}{\lambda}$) Using Watson's $\langle J_L \rangle$ values for Co^{++}, $f^{s}_{Co^{++}}$ is plotted against $\frac{\sin \theta}{\lambda}$ in the Figure 3 It is found to be in agreement within 6 % with the experimentally determined form factor for Co^{++}, The free ion form factor $\langle J_0 \rangle$ is also plotted for comparison The spherically symmetric form factor calculated with $|\Psi_{Co^{++}}\rangle$, i e, (8), as the ground state wavefunction of Co^{++}, is found to be

Figure 3

Plot of the theoretically determined spherically symmetric magnetic form factor for Co^{++} vs $(\sin\theta)/\lambda$ For comparison the experimental form factor and the free-ion form factor $\langle J_0 \rangle$ for Co^{++} are plotted



$$f^{s'}_{Co^{++}}(K) = \langle J_0 \rangle + 0\ 362\ \langle J_2 \rangle + 0\ 161\ \langle J_4 \rangle \qquad (38)$$

which almost coincides with $f^{s}_{Co^{++}}(K)$ This endorses the arguments, given in section 3, in the favour of the use of $|\psi_{Co^{++}}\rangle$

2 5 2 <u>For Fe^{++} in FeO</u>

There are four holes in the 3d shell of Fe^{++} The ^5D functions occurring in (27) are expressed in terms of these four hole wavefunction in appendix 3b Since we are concerned with the spherically symmetric part of the form factor, we shall omit $\langle \psi_{Fe^{++}}|H_x|\psi_{Fe^{++}}\rangle$ and $\langle \psi_{Fe^{++}}|H_y|\psi_{Fe^{++}}\rangle$ terms in (31) (appendix 2) Now by straightforward simplification we obtain

$$\frac{\langle \psi_{Fe^{++}}|H_Z|\psi_{Fe^{++}}\rangle}{\left[\langle \psi_{Fe^{++}}|H_Z|\psi_{Fe^{++}}\rangle\right]_{K=0}}$$

$$= \frac{1}{2\ 212}\Big[(0\ 909)^2\ \langle 2|e^{i\vec{K}\ \vec{r}}|2\rangle + \frac{1}{2}\langle 1|e^{i\vec{K}.\vec{r}}|1\rangle + \frac{1}{2}\langle 0|e^{i\vec{K}.\vec{r}}|0\rangle$$

$$+ \frac{1}{2}\langle 1|f(\vec{K}\ \vec{r})|1\rangle + (0\ 395)^2\ \langle 2|f(\vec{K}.\vec{r})|2\rangle$$

$$+ (0\ 135)^2\ (-\tfrac{1}{2})\ \langle 1|f(\vec{K}\ \vec{r})|1\rangle\Big] \qquad (39)$$

Using the results of appendix 4, (39) becomes

$$\frac{1}{2\ 212}\Big[1\ 652\ \langle J_0\rangle + 0\ 560\ \langle g_0\rangle$$

$$+ Y_{20}(K)\ (\tfrac{5\pi}{7})^{1/2}\ (0\ 826\ \langle J_2\rangle - 0\ 184\ \langle g_2\rangle)$$

$$+ Y_{40}(K)\ \tfrac{\sqrt{\pi}}{7}\ (3\ 305\ \langle J_4\rangle - 2.920\ \langle g_4\rangle)\Big] \qquad (40)$$

Using the explicit forms of $Y_{20}(\hat{K})$ and $Y_{40}(\hat{K})$ in (40) from appendix 5, we can separate out the spherical form factor,

$$f^{s}_{Fe^{++}}(K) = \frac{1}{2\ 212}\left[1\ 652\ \langle J_0\rangle + 0\ 560\ \langle g_0\rangle - \frac{5}{28}(0\ 326\ \langle J_2\rangle - 0\ 184\ \langle g_2\rangle) + \frac{9}{112}(3\ 305\ \langle J_4\rangle - 2\ 920\ \langle g_4\rangle)\right] \quad (41)$$

$$= \frac{1}{2\ 212}\left[1\ 652\ \langle J_0\rangle - 0\ 147\ \langle J_2\rangle + 0\ 265\ \langle J_4\rangle + 0\ 560\ (\langle g_0\rangle - \tfrac{1}{2}\langle g_2\rangle) + 0\ 313\ (\langle g_2\rangle - \tfrac{3}{4}\langle g_4\rangle)\right] \quad (42)$$

Now, using the relation (36) we have for (42)

$$f^{s}_{Fe^{++}} = \langle J_0\rangle + 0\ 257\ \langle J_2\rangle + 0\ 190\ \langle J_4\rangle \quad (43)$$

Using the values of $\langle J_0\rangle$, $\langle J_2\rangle$ and $\langle J_4\rangle$ as tabulated by Watson,[19] $f^{s}_{Fe^{++}}$ is plotted against $\sin\theta/\lambda$ in Figure 4 For comparison the free ion form factor $\langle J_0\rangle$ is also plotted Unlike CoO, no experimental results of neutron scattering by vacancy free FeO are available for comparison with the present theoretical form factor

2 6 CONCLUSION

As shown in Figure 4, the 11 % of 15-17 % expansion of the experimental form factor for Co^{++} in CoO from that of the free Co^{++} ion can be explained by inclusion of the orbital effect There are two small corrections which must also be considered First, the covalency effect which was studied by Hubbard and Marshall[20] for Mn^{++}, Ni^{++} etc in the strong field

Figure 4

Plot of theoretically predicted spherically symmetric magnetic form factor for Fe^{++} vs $(\sin\theta)/\lambda$ For comparison, the form factor for the free Fe^{++} ion, i e , $\langle J_o \rangle$, is plotted

representation using the 'spin only' form factor Such studies are difficult for CoO and FeO due to degenerate ground state and sizeable residual orbital moments Secondly, the spin polarization of fully occupied shells as discussed by Watson and Freeman[21] was found to expand the form factor of Ni^{++} by 4 % relative to the 'restricted' Hartree Fock calculation A similar expansion in the form factor of Co^{++} is expected Thus, the 4-6 % discrepancy in the theoretical and the experimental form factor for Co^{++} necessitates the quantitative estimation of the above mentioned corrections, which is out of the scope of the present work

APPENDIX 1

If the magnetic moment $\vec{\mu}_j$ of the j^{th} ion is oriented at (θ_j , ϕ_j) with respect to the crystal fixed coordinate system (Fig 5), the corresponding rotation operator R_j is given by

$$\begin{aligned} R_j &= R(\theta_j , \phi_j) \\ &= R_L(\theta_j , \phi_j) R_S(\theta_j , \phi_j) \\ &= e^{-i\phi_j L_Z} e^{-i\theta_j L_Y} e^{-i\phi_j S_Z} e^{-i\theta_j S_Y} \end{aligned} \tag{A1}$$

Clearly, $R(\theta_j , \phi_j)$ is arbitrary by a rotation about $\vec{\mu}_j$. Now, the rotation matrix corresponding to (A1) in ordinary space is

$$R^{(3)}(\theta_j , \phi_j) = R_l^{(3)}(\theta_j , \phi_j) R_S^{(3)}(\theta_j , \phi_j) \tag{A2}$$

where

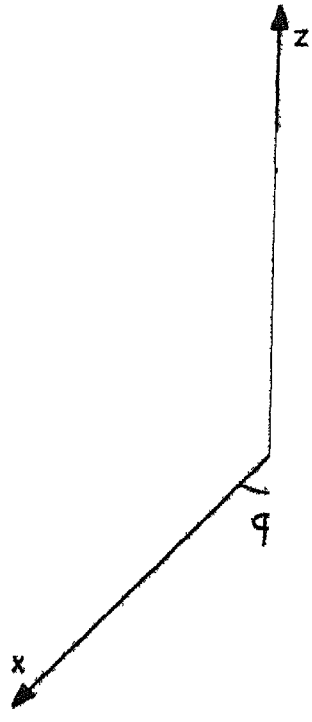
$$R_l^{(3)}(\theta_j , \phi_j) = \begin{pmatrix} \cos\theta_j \cos\phi_j & -\sin\phi_j & \sin\theta_j \cos\phi_j \\ \cos\theta_j \sin\phi_j & \cos\phi_j & \sin\theta_j \sin\phi_j \\ -\sin\theta_j & 0 & \cos\theta_j \end{pmatrix} \tag{A3}$$

is the rotation matrix for orbital moments, and a rotation matrix identical to (A3) can be written for spin moments as well

Now, we have for the spin part of (16),

Figure 5

Orientation of the magnetic moment of the j^{th} ion in the crystal-fixed coordinate system θ_j and ϕ_j are polar and azimuthal angles of the magnetic moment $\vec{}_j$



PRBL/Nov/BFlux/f

FIGURE 5

$$\mathcal{R}^{-1}_{S_J} S_x \mathcal{R}_{S_J} = \sum_1 R^{(3)}_{S_{11}} S_1$$

$$= S_x \cos\theta_J \cos\phi_J - S_y \sin\phi_J + S_z \sin\theta_J \cos\phi_J \qquad (A4)$$

Indices 1, 2 and 3 denote x, y and z components respectively

$$\mathcal{R}^{-1}_{S_J} S_y \mathcal{R}_{S_J} = \sum_1 F^{(3)}_{S_{21}} S_1$$

$$= S_x \cos\theta_J \sin\phi_J + S_y \cos\phi_J + S_z \sin\theta_J \sin\phi_J \qquad (A5)$$

and,

$$\mathcal{R}^{-1}_{S_J} S_z \mathcal{R}_{S_J} = \sum_1 R^{(3)}_{S_{31}} S_1$$

$$= -S_x \sin\theta_J + S_z \cos\theta_J \qquad (A6)$$

and similar expressions for $\mathcal{R}_1^{-1} \vec{I} \mathcal{R}_1$ can be written down
It can be readily seen by using the explicit form of $|\psi_{Co^{++}}\rangle$ given in Appendix 2a, that

$$\langle\psi_{Co^{++}}|\sum_J e^{i\vec{K}\vec{r}_J} S_{J_x}|\psi_{Co^{++}}\rangle = \langle\psi_{Co^{++}}|\sum_J e^{i\vec{K}\vec{r}_J} S_{J_y}|\psi_{Co^{++}}\rangle = 0 \qquad (A7)$$

Now,

$$\langle q_1|\sum_J e^{i\vec{K}\vec{r}_J} \vec{S}_J|q_1\rangle \ , \quad 1 = 1,2,3,4$$

$$= \langle\psi_{Co^{++}}| \sum_j e^{i\vec{K}\vec{r}_j} \mathcal{R}_{l}^{-1} \vec{S}_j \mathcal{R}_{l} |\psi_{Co^{++}}\rangle$$

$$= \langle\psi_{Co^{++}}| \sum_j e^{i\vec{K}\vec{r}_j} \mathcal{R}_{S_l}^{-1} \vec{S}_j \mathcal{R}_{S_l} |\psi_{Co^{++}}\rangle$$

$$= \langle\psi_{Co^{++}}| \sum_j e^{i\vec{K}\vec{r}_j} (\hat{x} S_{j_z} \sin\theta_l \cos\phi_l + \hat{y} S_{j_z} \sin\theta_l \sin\phi_l + \hat{z} S_{j_z} \cos\theta_l)|\psi_{Co^{++}}\rangle$$

$$= \hat{K}_l \langle\psi_{Co^{++}}| \sum_j e^{i\vec{K}\vec{r}_j} S_{j_z} |\psi_{Co^{++}}\rangle \qquad (A8)$$

where K_l is the unit vector in the direction of the magnetic moment of the l^{th} ion For the orbital part of $\vec{n}$ viz , $\frac{1}{4}\sum_l(\vec{l}_l f_l + f_l \vec{l}_l)$, we can proceed parallel to the spin part, but the difference is that

$$\langle\psi_{Co^{++}}| \frac{1}{4} \sum_j (l_{j_x} f_j + f_j l_{j_x}) |\psi_{Co^{++}}\rangle \neq 0$$

and,

$$\langle\psi_{Co^{++}}| \frac{1}{4} \sum_j (l_{j_y} f_j + f_j l_{j_y}) |\psi_{Co^{++}}\rangle \neq 0$$

So,

$$\langle q_l| \sum_j \frac{1}{4} (\vec{l}_j f_j + f_j \vec{l}_j) |q_l\rangle$$

$$= \langle\psi_{Co^{++}}| \mathcal{R}_{l_l}^{-1} \left(\sum_j \frac{1}{4}(\vec{l}_j f_j + f_j \vec{l}_j) \right) \mathcal{R}_{l_j} |\psi_{Co^{++}}\rangle$$

$$= \hat{K}_l \langle\psi_{Co^{++}}| \frac{1}{4} \sum_j (l_{j_z} f_j + f_j l_{l_z}) |\psi_{Co^{++}}\rangle$$

$$+K_{l'} \langle\psi_{Co^{++}}| \frac{1}{4} \sum_j (l_{j_x} f_j + f_j l_{j_x}) |\psi_{Co^{++}}\rangle$$

$$+K_{l''} \langle\psi_{Co^{++}}| \frac{1}{4} \sum_j (l_{j_y} f_j + f_j l_{j_y}) |\psi_{Co^{++}}\rangle \qquad (A9)$$

where,

$$\hat{K}_1 = \hat{x} \operatorname{Sin} \theta_1 \operatorname{Cos} \phi_1 + \hat{y} \operatorname{Sin} \theta_1 \operatorname{Sin} \phi_1 + \hat{z} \operatorname{Cos} \theta_1$$

$$\hat{K}_{1'} = \hat{x} \operatorname{Cos} \theta_1 \operatorname{Cos} \phi_1 + \hat{y} \operatorname{Cos} \theta_1 \operatorname{Sin} \phi_1 - \hat{z} \operatorname{Sin} \theta_1$$

$$\hat{K}_{1''} = - \hat{x} \operatorname{Sin} \phi_1 + \hat{y} \operatorname{Cos} \phi_1$$

which are unit vectors in the direction of new z, x and y axes respectively

APPENDIX 2

$\langle q_f| \frac{1}{4} \Sigma_l (l_{l_x} f_l + f_l l_{l_x}) |q_f\rangle$ and $\langle q_f| \frac{1}{4} \Sigma_l (l_{l_y} f_l + f_l l_{l_y}) |q_f\rangle$ (where $|q_f\rangle = |\Psi_{Co^{++}}\rangle$ or $|q_f\rangle = |\Psi_{Fe^{++}}\rangle$), will consist of the matrix elements of the following type

$$\langle dm| f(\vec{K}.\vec{r}) |dm'\rangle \qquad \text{(A10)}$$

with

$$|m - m'| = 1$$

It is obvious from (A11) that only $Y^*_{L,\pm 1}(K)$, (L = 2 and 4) will occur in (A10), and as shown in the Appendix 5, these do not contribute to the spherical symmetric part

APPENDIX 3a

Since $|\psi_{Co^{++}}>$, i e , (9) is made up of $|^4T>$ and $|^4P>$, we expand all the wavefunctions occurring in these states in terms of three hole functions of d shell as follows[22]

^4F state function

$$|M_S = \tfrac{3}{2} , M_L = 3> = (2^+, 1^+, 0^+)$$

$$|M_S = \tfrac{3}{2} , M_L = -1> = \frac{1}{\sqrt{10}} \left[\sqrt{6}\ (1^+, 0^+, -2^+) + 2(2^+, -1^+, -2^+)\right]$$

$$|M_S = \tfrac{1}{2} , M_L = 0 > = \frac{1}{\sqrt{15}} \left[(1^-, 0^+, -1^+) + (1^+, 0^-, -1^+) + (1^+, 0^+, -1^-) + 2\ (2^-, 0^+, -2^+) + (2^+, 0^-, -2^+) + (2^+, 0^+, -2^-)\right]$$

$$|M_S = -\tfrac{1}{2}, M_L = -3> = \frac{1}{\sqrt{3}}\left[(0^-, -1^-, -2^+) + (0^+, -1^-, -2^-) + (0^-, -1^+, -2^-)\right]$$

$$|M_S = -\tfrac{1}{2}, M_L = 1> = \frac{1}{\sqrt{30}} \left[\sqrt{6}\ (2^-, 0^-, -1^+) + (2^-, 0^+, -1^-) + (2^+, 0^-, -1^-) + 2\ (2^-, 1^-, -2^+) + (2^-, 1^+, -2^-) + (2^+, 1^-, -2^-)\right]$$

^4P state functions

$$|M_S = \tfrac{3}{2} , M_L = -1> = \frac{1}{\sqrt{10}} \left[-2(1^+, -2^+, 0^+) + \sqrt{6}(2^+, -2^+, -1^+)\right]$$

$$|M_S = \tfrac{1}{2} , M_L = 0 > = \frac{1}{\sqrt{15}} \left[-2\ (1^+, -1^-, 0^+) + (1^-, -1^+, 0^+) + (1^+, -1^+, 0^-) + (2^-, -2^+, 0^+) + (2^+, -2^-, 0^+) + (2^+, -2^+, 0^-)\right]$$

$$|M_S = -\frac{1}{2}\,,\ M_L = 1\rangle = \frac{1}{\sqrt{30}}\Big[-2\ (2^-,-1^-,0^+) + (2^+,-1^-,0^-) + (2^-,-1^+,0^-) + \sqrt{6}\ (2^-,-2^-,1^+) + (2^-,-2^+,1^-) + (2^+,-2^-,1^-)\Big]$$

Since $l = 2$ and $s = \frac{1}{2}$ for each single particle wavefunction, only m_l and m_s are specified + and - superscripts over m_l indicate $m_s = \frac{1}{2}$ and $-\frac{1}{2}$ respectively Parenthesis denotes the antisymmetric product function, i e , Slater determinant made up of the single particle functions given inside it

APPENDIX 3b

To express $|\Psi_{Fe^{++}}\rangle$, i e , (27) in terms of single particle wavefunctions (there are four holes in the 3d shell of Fe^{++}) we need the expansion of ^5D states as follows[22]

$$|M_S = 2,\ M_L = -1\rangle = (2^+,0^+,-2^+,-1^+)$$

$$|M_S = 1,\ M_L = 2\rangle = \frac{1}{2}\left[(2^-,1^+,-1^+,0^+) + (2^+,1^-,-1^+,0^+) + (2^+,1^+,-1^-,0^+) + (2^+,1^+,-1^+,0^-)\right]$$

$$|M_S = 1,\ M_L = -2\rangle = \frac{1}{2}\left[(1^-,0^+,-2^+,-1^+) + (1^+,0^-,-2^+,-1^+) + (1^+,0^+,-2^-,-1^+) + (1^+,0^+,-2^+,-1^-)\right]$$

$$|M_S = 0,\ M_L = 1\rangle = \frac{1}{\sqrt{6}}\left[(2^-,1^-,-2^+,0^+) + (2^-,1^+,-2^-,0^+) + (2^-,1^+,-2^+,0^-) + (2^+,1^-,-2^-,0^+) + (2^+,1^-,-2^+,0^-) + (2^+,1^+,-2^-,0^-)\right]$$

The notation is the same as in Appendix 3a

APPENDIX 4

We have the following relation,[8,2]

$$\langle dm|\ \bar{\Phi}(\vec{K}\,\vec{r})|dm'\rangle$$

$$= 4\pi \sum_{LM} i^{L} \langle h_L\rangle Y^{*}_{LM}(K) \int Y^{*}_{2m}(r)\, Y_{LM}(r)\, Y_{2m'}(r)\, d\Omega$$

$$= \sum_{L} 2[(2L+1)\pi]^{1/2} i^{L} \langle h_L\rangle C^{L}(2,m,2,m')\, Y^{*}_{Lm-m'}(\hat{K}) \qquad (A11)$$

where $\langle h_L\rangle = \int_0^\infty R^2(r)\, h_L(Kr) r^2 dr$, and the coefficients $C^L(l,m,l'm')$ are tabulated by Condon and Shortley[22] (nonzero terms in (A11) are for $L = 0$, 2 and 4 only). If $\bar{\Phi}(\vec{K}\,\vec{r}) = e^{i\vec{K}\,\vec{r}}$, $h(Kr)$ is the spherical Bessel function $j_L(Kr)$, while for $\bar{\Phi}(\vec{K}\,\vec{r}) = f(\vec{K}\,\vec{r})$, $h_L(Kr) = g_L(Kr)$,

where $g_L(\vec{K}\,\vec{r}) = \frac{1}{2} i^{-L} \int_{-1}^{+1} f(\vec{K}\,\vec{r}\,\mu)\, P_L(\mu)\, d\mu$,

$P_L(\mu)$ are Legendre polynomials

The following matrix elements are needed

$$\langle 2,2|\ \bar{\Phi}(\vec{K}.\vec{r})|2,2\rangle$$

$$= \langle 2,-2|\ \bar{\Phi}(\vec{K}.\vec{r})|2,-2\rangle$$

$$= \langle h_o\rangle + \frac{4}{7}(5\pi)^{1/2} Y_{20}(K) \langle h_2\rangle + \frac{2\sqrt{\pi}}{7} Y_{40}(K) \langle h_4\rangle, \qquad (A12)$$

$$\langle 2,1|\ \bar{\Phi}(\vec{K}\,\vec{r})|2,1\rangle$$

$$= \langle 2,-1|\ \bar{\Phi}(\vec{K}.\vec{r})|2,-1\rangle$$

$$= \langle h_o\rangle - \frac{2}{7}(5\pi)^{1/2} Y_{20}(K) \langle h_2\rangle - \frac{8\sqrt{\pi}}{7} Y_{40}(K) \langle h_4\rangle, \qquad (A13)$$

$$\langle 2,0|\ \underline{\Phi}(\vec{r}\ \vec{r})|2,0\rangle$$

$$= \langle h_0\rangle - \frac{4}{7}(5\pi)^{1/2} Y_{20}(\hat{r})\langle h_2\rangle + \frac{4}{7}(9\pi)^{1/2} Y_{40}(\hat{r})\langle h_4\rangle, \tag{A14}$$

We have used the fact that $Y_{20}(\hat{r})$ and $Y_{40}(\hat{r})$ are real

APPENDIX 5

We have,[23]

$$Y_{oo}(K) = \left(\frac{1}{4\pi}\right)^{1/2} \tag{A15}$$

$$Y_{20}(K) = \left(\frac{5}{4\pi}\right)^{1/2} \left(\frac{1}{4}\right)^{1/2} \frac{3K_z^2 - K^2}{K^2} \tag{A16}$$

$$Y_{40}(K) = \left(\frac{9}{4\pi}\right)^{1/2} \left(\frac{1}{64}\right)^{1/2} \frac{35 K_z^4 - 30 K_z^2 K^2 + 3K^4}{K^4} \tag{A17}$$

$$Y_{2\pm1}(K) = \mp \left(\frac{15}{8\pi}\right)^{1/2} \frac{(K_x \pm iK_y) K_z}{K^2} \tag{A18}$$

$$Y_{4\pm1}(K) = \mp \left(\frac{9}{4\pi}\right)^{1/2} \left(\frac{5}{16}\right)^{1/2} \frac{(K_x \pm iK_y)}{K} \times \frac{(7K_z^3 - 3K^2 K_z)}{K^3} \tag{A19}$$

Clearly, the spherical parts of (A15), (A16) and (A17) is as follows,

$$\left[Y_{oo}(K)\right]_{sph} = \left(\frac{1}{4\pi}\right)^{1/2} ,$$

$$\left[Y_{20}(K)\right]_{sph} = -\frac{1}{4}\left(\frac{5}{\pi}\right)^{1/2} ,$$

$$\left[Y_{40}(K)\right]_{sph} = \frac{9}{16\sqrt{\pi}} ,$$

while the spherical part of (A18) and (A19) is zero

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SECTION - B

INELASTIC SCATTERING OF NEUTRONS

CHAPTER - III

SCATTERING CROSS-SECTION FOR FERROMAGNETIC RARE-EARTH CRYSTALS

3 1 INTRODUCTION

We consider a ferromagnetic rare-earth crystal consisting of N magnetic atoms, each of which has total angular momentum $\bar{J}$ and coupled to the others by the isotropic exchange interaction,

$$H = -\sum_{l \neq J} I(\bar{R}_l - \bar{R}_J)\, \bar{J}_l\, \bar{J}_J \qquad (1)$$

where R_l and R_J denote the lattice sites

For temperatures much lower than Curie point the magnetization is very close to the saturation magnetization at absolute zero Hence the average projections of total angular momentum of the ions, on the direction of spontaneous magnetic moment differ little from $|\bar{J}|$ Then total angular momentum operators are approximately expressed in terms of Bose operators as follows

$$b_J = \frac{1}{\sqrt{2J}} J_J^+$$

$$b_J^+ = \frac{1}{\sqrt{2J}} J_J^- \qquad (2)$$

$$b_J^+ b_J = J - J_z$$

It is useful to transform from b_J , b_J^+ to b_k , b_k^+ as follows

$$b_J = \frac{1}{\sqrt{N}} \sum_k e^{i\underline{k}\,\underline{R}_J} b_k \,,$$
$$b_J^+ = \frac{1}{\sqrt{N}} \sum_k e^{-i\underline{k}\,\underline{R}_J} b_k^+ \qquad (3)$$

Using (2) and (3) in (1), it can be shown that

$$H = \sum_k \epsilon_k b_k^+ b_k + \text{constant} \qquad (4)$$

where,
$$\epsilon_k = 2J \sum_{\underline{R}} I(\underline{R}) \quad (1 - e^{i\underline{k}\,\underline{R}}) \qquad (5)$$

3 2 CROSS-SECTION FOR ONE SUBLATTICE CASE

If electronic spin and orbital momentum both are responsible for the scattering of neutrons, it can be readily shown that the corresponding differential scattering cross-section in Born approximation for unpolarised neutrons is

$$\frac{d^2\sigma}{d\Omega\, dE_{p'}} = (r_o \gamma)^2 \frac{p'}{p} \sum_{J,J'} e^{-i\,\underline{q}\,(\underline{R}_J - \underline{R}_{J'})} \sum_{\alpha,\beta} (\delta_{\alpha\beta} - e_\alpha e_\beta)$$
$$\times \frac{1}{2\pi\hbar} \int_{-\infty}^{\infty} e^{\frac{it}{\hbar}(E_{p'} - E_p)} \langle P_J^\alpha(0)\, P_{J'}^\beta(t)\rangle dt \qquad (6)$$

where,

$$\bar{P}_J = \sum_\nu^{Z_1} \left\{ e^{i\underline{q}\,\underline{r}_\nu} \underline{s}_\nu + \frac{1}{4\hbar} \left[\underline{l}_\nu f(\underline{q}\,\underline{r}_\nu) + f(\underline{q}\,\underline{r}_\nu)\, \underline{l}_\nu \right] \right., \qquad (7)$$

$\underline{q}$ is the scattering vector, $\underline{p}$ and $\underline{p}'$ are respectively the initial and the final wave vectors of the neutron, $\gamma = -1.91$, $r_o = \frac{e^2}{m_e c^2}$, the classical electron radius, $\underline{e} = \frac{\bar{q}}{|\bar{q}|}$, E_p and $E_{p'}$ are the energies corresponding to p and p' respectively

Also,

$$\langle P_j^{\alpha}(0)\ P_{j'}^{\beta}(t)\rangle = \frac{\mathrm{Trace}\left\{ e^{-H\beta'}\ P_j^{\alpha}(0)\ P_{j'}^{\beta}(t)\right\}}{\mathrm{Trace}\ \mathrm{Trace}\left\{ e^{-H\beta'}\right\}} \qquad (8)$$

where, $\beta' = \frac{1}{K_B T}$, $K_B = 1\ 380 \times 10^{-16}$ ergs/^{o}A (Boltzmann constant)

It follows from (8) that

$$\langle P_j^{\alpha}(0)\ P_{j'}^{\beta}(t)\rangle = \sum_{a,a'} e^{-E_a\beta'} \langle a|P_j^{\alpha}(0)|a'\rangle \langle a'|P_{j'}^{\beta}(t)|a\rangle \times \frac{\exp\left[\frac{it}{\hbar}(E_{a'}-E_a)\right]}{\sum_a e^{-E_a\beta'}} \qquad (9)$$

where $|a\rangle$ and $|a'\rangle$ are the scatterer states, i.e., eigen states of the Hamiltonian (1). Hence,

$$|a\rangle = |n_{k_1}, n_{k_2}, \ldots, n_{k_N}\rangle = \prod_i \frac{(b_{k_i}^{+})^{n_{k_i}}}{(n_{k_i}!)^{1/2}} |0\rangle \qquad (10)$$

where, $|0\rangle$ represents the vacuum state for magnons. This is also the ground state of the Hamiltonian (1) and corresponds to perfect alignment

$$|0\rangle = |J,J\rangle\ |J,J\rangle \ldots |J,J\rangle \qquad (11)$$

Transforming b_k^+ into b_j by (3), we obtain for (10) as

$$|a\rangle = \prod_{\iota} \left\{ \frac{1}{(l_1')^{1/2}\, N^{l/2}} \sum_{j_{11}, j_{21}, \ldots, j_{l1}} e^{i\underline{k}_1 (\underline{R}_{j_{11}} + \underline{R}_{j_{21}} + \cdots + \underline{R}_{j_{l1}})} \right.$$

$$\times b^+_{j_{11}} b^+_{j_{2i}} \cdots b^+_{j_{l1}} |0\rangle \qquad (12)$$

$$l_1 = n_{k_1} \rightarrow \text{in language}$$

Using (2),

$$|a\rangle = \prod_{\iota} \frac{1}{(l')^{1/2}\, (2jN)^{l/2}} \sum_{j_{11}, j_{21}, \ldots, j_{l1}} e^{i\underline{k}_1 (\underline{R}_{j_{11}} + \underline{R}_{j_{21}} + \cdots + \underline{R}_{j_{l1}})}$$

$$\times J^-_{j_{11}} J^-_{j_{21}} \cdots J^-_{j_{l1}} |0\rangle \qquad (13)$$

$$l = n_{k_1}$$

Thus we transformed the magnon states as a superposition of crystal states given by the product of individual ionic wave-functions This is done with a view to simplify the matrix elements occurring in (9) An alternate approach would have been to transfer $\bar{P}_j$ to $\bar{J}_j$ and then to magnon variables But this is not possible for arbitrary values of $\bar{q}$

3.3 CALCULATION OF THE MATRIX ELEMENTS

Substituting for $|a\rangle$ and $|a'\rangle$ from (13) ($|a'\rangle$ is obtained from (13) by replacing n_{k_1} by n'_{k_1}), we get,

$$\langle a|P_J^{\alpha}|a'\rangle$$

$$= \langle 0| \prod_{l=1}^{N} \frac{1}{(l!\, l'!)^{1/2}\, (2JN)^{\frac{l+l'}{2}}} \sum_{J_{1l}\, J_{2l}} \sum_{J_{1l}, J'_{1l}, J'_{2l}, \ldots, J'_{l'1}} e^{-ik_l (\underline{R}_{J_{1l}} + \underline{R}_{J_{2l}} + \cdots + \underline{R}_{J_{1l}})} \times e^{ik_l (\underline{R}_{J'_{1l}} + \underline{R}_{J'_{2l}} + \cdots + \underline{R}_{J'_{l'1}})}$$

$$\times (J^+_{J_{1l}} J^+_{J_{2l}} \cdots J^+_{J_{1l}})_{l=n_{k_l}} P_J^{\alpha} \prod_{l=1}^{N} (J^-_{J'_{l'1}} J^-_{J'_{2l}} J^-_{J'_{1l}})_{l'=n'_{k_l}} |0\rangle$$

$$l'=n'_{k_l} \qquad (14)$$

The matrix element occuring in (14) is

$$\langle 0| \prod_{l=1}^{N} (J^+_{J_{1l}} J^+_{J_{2l}} \cdots J^+_{J_{1l}})_{l=n_{k_l}} P_J^{\alpha} \prod_{l=1}^{N} (J^-_{J'_{l'1}} J^-_{J'_{2l}} J^-_{J'_{1l}})_{l'=n'_{k_l}} |0\rangle \qquad (15)$$

Since P_J^{α} depends only on J^{th} ion and $|0\rangle$ is given by (11), we can split up (15) in product of individual ionic parts

Taking $\sum_l n_{k_l} = n$ and $\sum_l n'_{k_l} = n'$, we consider the following cases

(1) $n = n'$

Then in order that (15) may not vanish, we must have

$$\prod_{l=1}^{N} (J^+_{J_{1l}} J^+_{J_{2l}} \cdots J^+_{J_{1l}})_{l=n_{k_l}} \equiv \prod_{l=1}^{N} (J^+_{J'_{l'i}} J^+_{J'_{2l}} J^+_{J'_{1l}})_{l'=n'_{k_l}} \qquad (16)$$

Otherwise orthogonality relations for individual ionic wavefunctions will make (15) vanish Moreover not more than 2J of $J^{+}_{m_1}$ may be equal on either sides

(2) $n > n'$, $(n-n') = r$

Then in order that (15) may not vanish, a condition similar to (16) must hold for n' out of n $J^{-}_{m_1}$ and the remaining r of $J^{+}_{m_1}$ must belong to the j^{th} ion so as to occur in matrix element with P^{α}_{j} Moreover $r < 2J$ must be satisfied otherwise the r of $J^{+}_{m_1}$ acting on j^{th} ionic wavefunction will make it zero

(3) $n < n'$, $n' - n = r$

The conditions of nonvanishing of (15) are similar to those for the case (2)

Now, we shall simplify (14) for $n>n'$ The cases $n<n'$ and $n = n'$ will immediately follow from this

So, for $n>n'$ we have from (14),

$$\sum_{a'} \langle a \mid P^{\alpha}_{j} \mid a' \rangle_{n-n'=r}$$

$$= \langle 0 \mid \sum_{\{r_1\}} \prod_{1=1}^{N} \left\{ \sqrt{n_{k_1}(n_{k_1}-1)\cdots(n_{k_1}-r_1+1)} \times \frac{1}{(2JN)^{\frac{r_1}{2}} + n'_{k_1}} \right.$$

with $\sum_{i} r_1 = r$

$$\left. \times \sum_{J_{11}, J_{21}, \cdots J_{1'1}} e^{-i r_1 (\underline{k}_1 \underline{R}_j)} (J^{+}_{J_{11}} J^{+}_{J_{21}} \cdots J^{-}_{J_{1'1}}) \right\}$$

$1' = n'_{k_1}$

$$(J_j^+)^{r_1} P_J^{\alpha} \prod_{1=1}^{N} \left\{ J^-_{J_{1'1}} \quad J^-_{J_{21}} J^-_{J_{11}} \right\}_{1'=n'_{k_1}} |0\rangle + 0\left(\frac{n}{N}\right) \tag{17}$$

where, $n_{k_1} - n'_{k_1} = r_1$ and $\{r_1\} \equiv r_1, r_2, \ldots, r_N$

Hence, $n - n' = \sum_1 r_1 = r$

Use is made of the fact that (17) will be nonzero for

$$J^+_{J_{11}} J^+_{J_{21}} \cdot J^+_{J_{1'1}} \equiv J^+_{J'_{11}} J^+_{J'_{21}} \quad J^+_{J'_{1'1}}$$

which is a more stringent condition than (16) and results due to occurrence of $\sum_{J_{11}} e^{i(\underline{k}_1 - \underline{k}_{1'}) \underline{R}_{J_{11}}} \sim \delta_{\underline{k}_1, \underline{k}'_1}$ which make (17) zero if $J^{\pm}_{J_{m1}}$ arising from different $\underline{k}_1$ happens to be identical The error involved in summing over all the values of J_{11} is of the order of $\frac{n}{N}$ i.e , very small as shown later on

Also, r_1 out of n_{k_1} can be chosen in $n_{k_1}(n_{k_1}-1) \ldots (n_{k_1}-r_1+1)$ ways and the remaining n'_{k_1} $(= n_{k_1} - r_1)$ are permuted in n'_{k_1} ways Thus, these factors occur in the numerator of (17) on opening the summations over $J_{11}, J_{21} \ldots J_{11}$ In order to further simplify (#7) we shall open the remaining summations one by one Each of the summation can be broken up as follows -

$$\sum_{J_{11}} \rightarrow \sum'_{J_{11}} + \sum''_{J_{11}} + \sum'''_{J_{11}} + (n' + 1) \text{ terms at the most}$$

where, $\Sigma'_{J_{1\iota}}$ excludes terms with $J_{1\iota}$ equal to $J_{2\iota}, J_{3\iota}, \quad J_{1'\iota}$ $(\iota = 1,2, \quad ,N)$

or ∅

$\Sigma''_{J_{1\iota}}$ contains only the terms with $J_{1\iota}$ equal to one of the $J_{2\iota}, J_{3\iota}, \quad ,J_{1'\iota},J$ $(\iota = 1,2, \quad ,N)$

$\Sigma'''_{J_{1\iota}}$ contains only the terms with $J'_{1\iota}$ equal to two of the $J_{2\iota},J_{3\iota}, \quad ,J_{1'\iota},J$ $(\iota = 1,2, \quad ,N)$

and so on

There are at the last $(N-n'-1)$ terms in $\Sigma'_{J_{1\iota}}$ and all these terms are identical

The number of terms in $\Sigma''_{J_{1\iota}}$ can be at the most $(n'+1)$, in $\Sigma'''_{J_{1\iota}}$ at the most $\frac{(n'+1)}{2}$, in $\Sigma^{IV}_{J_{1\iota}}$ at the most $\frac{(n')}{3}$ and so on

In spin wave approximation when $\frac{n'}{N} \ll 1$, we can safely assume that $\Sigma_{J_{1\iota}}$ has N identical terms each of which is same as that for $J_{1\iota}$ not equal to any of $J_{2\iota},J_{3\iota}, \quad ,J_{1'\iota}$ $(\iota=1, \quad N),J$

Similarly we can open $\Sigma_{J_{2\iota}}$, $\Sigma_{J_{3\iota}}$, and finally obtain under spin wave approximation,

$$\sum_{a'} \left[\langle a|P^{\alpha}_{J}|a'\rangle \right]_{n-n'=r}$$

$$= \langle 0 \Big| \sum_{\{r_{1\iota}\}} \prod_{\iota=1}^{N} \left\{ \sqrt{n_{k_\iota}(n_{k_\iota}-1) \quad (n_{k_\iota}-r_\iota+1)} \quad \times \right.$$

with restriction $\sum_{\iota} r_\iota = r$.

$$\times \frac{\overset{N}{\underset{l=1}{\prod}} n'_{k_l}}{(2JN)^{r_l/2}\ \overset{N}{\underset{l=1}{\prod}} n'_{k_l}!}\ e^{-i\, r_l(\underline{k}_l \cdot \underline{R}_j)}$$

$$\sim \{ J^+_{j_{11}} J^+_{j_{28}} \cdots J^+_{j_{1'1}} \} (J^+_j)^r P^\alpha_j \overset{N}{\underset{l=1}{\prod}} \{ J^-_{j_{1'1}} \cdots J^-_{j_{21}} J^-_{j_{11}} \} |0\rangle$$

$$l'=n'_{k_l} \qquad\qquad l'=n'_{k_l}$$

(18)

where none of the j_{ml} is equal to the others or j. It is easy to see that (18) is simplified to

$$\sum_{\{r_i\}} \overset{N}{\underset{l=1}{\prod}} \sqrt{n_{k_l}(n_{k_l}-1)\cdots(n_{k_l}-r_l+1)}\ \frac{1}{(2JN)^{r/2}}\ e^{-i\, r_l(\underline{k}_l \cdot \underline{R}_j)}$$

with $\sum_l r_l = r$

$$\times \sqrt{(2J)\ \{2(2J-1)\}\ \{3(2J-2)\}\cdots\{r(2J-r+1)\}}\ \langle J, J-r|P^\alpha_j|JJ\rangle \qquad (19)$$

To obtain $\sum_{a'} \langle a|P^\alpha_j|a'\rangle_{n'-n=r}$ from (19), take its complex conjugate and change n_{k_l} to n'_{k_l}. Again using $n_{k_l} = n'_{k_l} - r_l$, we obtain

$$\sum_{a'} \langle a|P^\alpha_j|a'\rangle_{n'-n=r}$$

$$\sim \sum_{\{r_l\}} \overset{N}{\underset{l=1}{\prod}} \sqrt{n'_{k_l}(n'_{k_l}-1)\cdots(n'_{k_l}-r_l+1)}\ \frac{1}{(2JN)^{r/2}}\ e^{i\, r_l(\underline{k}_l \cdot \underline{R}_j)}$$

with $\sum_i r_l = r$

$$\times \sqrt{(2J)\ 2(2J-1)\ 3(2J-2)\cdots r(2J-4+1)}\ \langle J, J|P^\alpha_j|J, J-r\rangle$$

$$\times \sqrt{(2J)\quad 2(2J-1)\quad 3(2J-2)\quad \cdots\quad r(2J-r+1)}\ \langle J,J|P_j^\alpha|J,J-r\rangle$$

$$= \sum_{r_1} \prod_{l=1}^{N} \sqrt{(n_{k_l}+1)(n_{k_l}+2)\cdots(n_{k_l}+r_l)}\ \frac{1}{(2JN)^{r/2}}\ e^{i\,r_l(\underline{k}_l\,\underline{R}_j)}$$

$$\text{with } \sum_l r_l = r$$

$$\times \sqrt{2J\{2(2J-1)\,,\,3(2J-2)\}\cdots\{r(2J-r+1)\}}\ \langle J,J|P_j^\alpha|J,J-r\rangle \qquad (20)$$

Also

$$\langle a|P_j^\alpha|a'\rangle_{n=n'}$$

$\simeq \langle J,J|P_j^\alpha|J,J\rangle$ which follows from (18) ~~or (20)~~ by putting r = 0 (21)

Now,

$$E_a = \sum_{l=1}^{N} \epsilon_{k_l} n_{k_l}\ ,\quad E_{a'} = \sum_{l=1}^{N} \epsilon_{k_l} n'_{k_l} \qquad (22)$$

$$\sum_a \equiv \sum_{\{n_{k_l}\}}\ ,\quad \sum_{a'} \equiv \sum_{\{n'_{k_l}\}} \quad \text{where } \{n_{k_l}\} \equiv n_{k_1}, n_{k_2}, \ldots, n_{k_N} \qquad (23)$$

$$\langle J,J|P_j^\alpha|J,J-r\rangle = \langle J,J|P^\alpha|J,J-r\rangle \qquad (24)$$

Eqn. (24) shows that matrix elements do not depend on the particular lattice site for which P_j^α is the interaction operator. This is true for one sublattice ferromagnetic crystal. Using (22) to (24) in (9) we obtain,

$$\langle P^{\alpha}_{J}(0)\, P^{\gamma}_{J'}(t)\rangle$$

$$= \sum_{\{n_{k_i}\}} \sum_{\{n'_k\}} e^{-\sum_{1}^{N} \epsilon_{k_1} n_{k_1} \beta'} \langle n_{k_1}, n_{k_2}, \ldots, n_{k_N} | P^{\alpha} | n'_{k_1}, n'_{k_2}, \ldots, n'_{k_N} \rangle$$

$$\times \langle n'_{k_1}, n'_{k_2}, \ldots, n'_{l_N} | P^{\beta} | n_{k_1}, n_{k_2}, \ldots, n_{l_N} \rangle$$

$$\times \langle \alpha\beta\, e^{it/\hbar \{ (\epsilon_{k_1} n'_{k_1} + \epsilon_{k_2} n_{k_2} + \cdots + \epsilon_{k_N} n_{k_N}) - (\epsilon_{k_1} n_{k_1} + \epsilon_{k_2} n_{k_2} + \cdots + \epsilon_{k_N} n_{k_N}) \}} \alpha\beta \rangle \Big/ \sum_{\{n_k\}} e^{-\beta' (\epsilon_{k_1} n_{k_1} + \epsilon_{k_2} n_{k_2} + \cdots + \epsilon_{k_N} n_{k_N})}$$

$$= \sum_{\{n_{k_i}\}} \sum_{\{n_k\}} e^{-\beta \sum_{1}^{N} \epsilon_{k_i} n_{k_i}} \langle \{n_{k_i}\} | P^{\alpha} | \{n'_k\} \rangle \langle \{n'_{k_i}\} | P^{\beta} | \{n_{k_i}\} \rangle \times$$

$$e^{\frac{it}{\hbar} \sum_{1}^{N} (\epsilon_{k_i} n'_{k_i} - \epsilon_k n_{k_i})} \Big/ \sum_{\{n_k\}} e^{-\beta' \sum_{1}^{N} \epsilon_{k_i} n_k} \qquad (25)$$

3 4 EXPRESSION FOR CROSS-SECTION

Using the relations

$$\Big| \sum_{J} e^{i\underline{q} \cdot \underline{R}_J} \Big|^2 = \frac{(2\pi)^3 N}{v_o} \sum_{\underline{z}} \delta(\underline{q} - \underline{z}) \,, \qquad (26)$$

$$\frac{1}{2\pi\hbar} \int_{-\infty}^{\infty} e^{\frac{i t E}{\hbar}} dt = \delta(E) \qquad (27)$$

and (19) to (21) as well as (25) in (6), we obtain

$$\frac{d^2\sigma}{d\Omega\, dE_{p'}} = (r_0\gamma)^2 \frac{p'}{p} \sum_{J,J'} e^{-i\underline{q}\cdot(\underline{R}_J-\underline{R}_{J'})} \sum_{\alpha,\beta} (\delta_{\alpha\beta}-e_\alpha e_\beta)$$

$$\times \frac{1}{2\pi\hbar}\int_{-\infty}^{\infty} dt\, e^{it/\hbar\,(E_{p'}-E_p)} \sum_{\{n_{k_1}\}} e^{-\beta'\sum_{1=1} \epsilon_{k_1} n_{k_1}} \Big/ \sum_{\{n_{k_1}\}} e^{-\beta'\sum_{1=1}^{N} \epsilon_{k_1} n_{k_1}}$$

$$\times \Bigg[\langle J,J|P^\alpha|J,J\rangle \langle J,J|P^\beta|JJ\rangle + \sum_{\{r_1\}} \prod_{1=1}^{N} \frac{1}{(2JN)^r}$$

$$\text{with } \sum_1 r_1 = r \neq 0$$

$$\times (2J)\ \{2(2J-1)\}\ \{3(2J-2)\} \qquad \{r(2J-r+1)\}$$

$$\langle JJ|P^\alpha|JJ-r\rangle \langle J,J-r|P^\beta|JJ\rangle \times$$

$$\Big\{ e^{i\gamma_i \underline{k}_i\cdot(\underline{R}_j-\underline{R}_{j'})} \times e^{\frac{it}{\hbar}\gamma_i \epsilon_K (n_{K_i}+1)(\gamma_K+2)} \qquad (n_K+\gamma_i)$$

$$+ e^{-i\gamma_i \underline{k}_i\cdot(\underline{P}\ \underline{P}_{j'})} \epsilon^{\frac{-it}{\hbar}\gamma_i \epsilon_{K_i}} n_K(n_K\ 1) \qquad (n_K-r+1)\Big\}\Bigg]$$

(28)

Hence,

$$\frac{d^2\sigma}{d\Omega\, dE_{p'}} = (r_0\gamma)^2\ e^{-2Wq} \sum_{\alpha,\beta} \langle JJ|P^\alpha|JJ\rangle \langle JJ|P^\beta|JJ\rangle (\delta_{\alpha\beta}-e_\alpha e_\beta)$$

$$\times \frac{(2\pi)^3 N}{v_0} \sum_{\underline{z}} \delta(\underline{q}-\underline{z})\ \delta(E_{p'}-E_p)$$

$$+ (r_o r')^2 \, e^{-2W_q} \sum_{\alpha\beta} (\delta_{\alpha\beta} - e_\alpha e_\beta) \sum_{p}' (1/N) \; \langle JJ|P^\alpha|J,J-1\rangle$$

$$\times \langle JJ-1|P^\beta|JJ\rangle \frac{(2\pi)^3}{V_o} N \sum_k \sum_{\underline{g}} \Big[(\langle n_k\rangle + 1) \, \delta(\underline{q} - \underline{k} - \underline{g}) \, \delta(E_{p'} - E_p + \epsilon_k)$$

$$+ \langle n_k \rangle \; \delta(\underline{q} + \underline{k} - \underline{g}) \, \delta(E_{p'} - E_p - \epsilon_k) \Big] \qquad (29)$$

where $\langle n_k \rangle = \dfrac{1}{e^{\beta \epsilon_k} - 1}$,

The factor $e^{-2W_q} = |\langle e^{i\underline{q}\,\underline{U}_J}\rangle|^2$ occurs due to lattice vibrations at finite temperature $\underline{U}_J$ is the displacement of j^{th} lattice site from the equilibrium position

Only terms upto one magnon scatterings have been retained in (29) The terms in (28) corresponding to r magnon scattering are smaller by a factor of $1/N^r$ than elastic scattering part Hence the contribution to the scattering cross section by more than one magnon scattering processes have been neglected in (29) It is interesting to note in the connection with (28) that in the frame-work of spin wave approximation, neutron inelastic scattering may occur due to absorption or emission of as many as 2J magnons This is a consequence of the interaction of neutron with orbital momentum of electrons, because under 'spin only' assumption only one magnon was responsible for the inelastic neutron scattering It follows from (29) that energy-momentum conservation laws for emission and absorption of magnon are identical as for 'spin only' case, but the intensities of

elastic and inelastic scattering peaks as well as their angular dependence are completely different To calculate the numerical value of (29), the following results can be used,

$$\langle JM|P_{q'}|JM'\rangle = \sqrt{4\pi} \sum_{K'Q'Q} \frac{2K'+1}{K'+1} A(K'-1, K') + B(K'-1, K') Y_{K'-1,Q}(\hat{q}) \langle K'-1,Q,K'Q'| 1q'\rangle \times \langle K'0' JM'|JM\rangle \quad (30)$$

where the expression for $A(K'-1, K')$ and $B(K'-1, K')$ are given by Lovesey and Rimmer Also,

$$P_0 = P^z \qquad P^z = P^0$$

$$P_1 = -\frac{1}{\sqrt{2}}(P^x + i P^y) \quad \text{or} \quad P^y = \frac{i}{\sqrt{2}}(P_1 + P_{-1}) \quad (31)$$

$$P_{-1} = \frac{1}{\sqrt{2}}(P^x - i P^y) \qquad P^x = \frac{P_{-1} - P_{-1}}{\sqrt{2}}$$

3.5 THE 'SPIN-ONLY' CASE

The expression for differential scattering cross section for 'spin only' case is obtained from (29) by taking

$$P^\alpha = F(\underline{q})\, S^\alpha$$

and $$|JM\rangle = |SM_S\rangle$$

Then

$$\sum_{\alpha\beta} (\delta\alpha\beta - e_\alpha e_\beta) \langle SS|S^\alpha|SS\rangle \langle SS|S^\beta|SS\rangle F^2(\underline{q}) = \left\{1 - (\bar{e}.\bar{m})^2\right\} F^2(\underline{q})\, S^2 \quad (32)$$

where $\bar{m}$ is unit vector along z axis

$$\sum_{\alpha\beta} (\delta\alpha\beta - e_\alpha e_\beta) \langle S,S|S^\alpha|S,S-1\rangle \langle S,S-1|S^\beta|S,S\rangle$$

$$= (1- e_x^2) \langle SS|S^x|SS-1\rangle \langle SS-1|S^x|SS\rangle$$

$$+ (1- e_y^2) \langle SS|S^y|SS-1\rangle \langle SS-1|S^y|SS\rangle$$

$$+ (1- e_x e_y) \{ \langle SS|S^x|SS-1\rangle \langle SS-1| S^y|SS\rangle$$

$$+ \langle SS|S^y|SS-1\rangle \langle SS-1|S^x|SS\rangle$$

$$= (1- e_x^2) \frac{1}{2\times 2}(+2S) + (1-e_y^2) \frac{1}{2\times 2} (+2S) + (1- e_x e_y) \left[2S \frac{1}{(\sqrt{2})^2} - \frac{2S\, i}{(\sqrt{2})^2} \right]$$

$$= + \frac{S}{2} (1 + e_z^2)$$

$$= + \frac{S}{2} (1 +(\bar{m}\, \bar{e})^2) \qquad (33)$$

We have used $S^x = \frac{S^+ + S^-}{2}$, $S^y = \frac{S^+ - S^-}{2i}$ in the above using (32) and (33) in (29) we get the same result as obtained for the 'spin only' case, i e

$$\frac{d^2\sigma}{d\Omega\, dE_{p'}} = (r_o \gamma)^2 F^2(\underline{q}) e^{-2W_q} S^2 \left\{ 1-(\bar{e}\, \bar{m})^2 \right\} \times \frac{(2\pi)^3 N}{v_o} \sum_{\underline{z}} \delta(\underline{q}-\underline{z})$$

$$\times \delta(E_{p'}-E_p) + (r_o \gamma)^2 F^2(\underline{q}) e^{-2W_q} \frac{S}{2} \frac{p'}{p} \left\{ 1+(\bar{e}\, \bar{m})^2 \right\}$$

$$\times \frac{(2\pi)^3}{v_o} \sum_{\underline{z}} \sum_{\underline{k}} \delta(\underline{q}-\underline{k}-\underline{z}) \delta(E_{p'}-E_p+ k) (1+\langle n_k\rangle)$$

$$+ \delta(\underline{q}+\underline{k}-\underline{z}) \delta(E_{p'}-E_p- k) \langle n_k\rangle \}$$

5 6 ALTERNATE APPROACH FOR FERROMAGNET (ONE SUB-LATTICE)

We have,

$$b_{k_1} = \frac{1}{\sqrt{2NJ}} \sum_{J_1} e^{i\underline{k}_1 \underline{R}_{J1}} J^+_{J_1}$$

$$= \frac{1}{\sqrt{2NJ}} \sum_{J_1 \neq J} e^{i\underline{k}_1 \cdot \underline{R}_{J_1}} J^+_{J_1} + \frac{1}{\sqrt{2NJ}} e^{i\underline{k}_1 \underline{R}_J} J^+_J$$

So,

$$\left[b_{k_1} , P^\alpha_j \right] = \frac{1}{\sqrt{2NJ}} e^{i\underline{k}_1 \underline{R}_J} \left[J^+_J, P^\alpha_J \right] \qquad (1)$$

because $\vec{P}_J$ is made up of operators for J^{th} ions alone hence commutes with J^+_{J1} for $J_1 \neq J$ i e with operators for other ions The assumption is that various ions are independent to each other. This is justified in molecular field approximation From (1),

$$b_{k_1} P^\alpha_J b_{k_1} + \frac{1}{\sqrt{2NJ}} e^{i\underline{k}_1 \cdot \underline{R}_J} \left[J^+_J, P^\alpha_J \right] \qquad (2)$$

$$b_{k_1} b_{k_1'} P^\alpha_j = P^\alpha_J b_{k_1} b_{k_1'} + \frac{1}{\sqrt{2NJ}} e^{i\underline{k}_1' \underline{R}_J} b_{k_1} \left[J^+_J, P^\alpha_J \right]$$

$$+ \frac{1}{\sqrt{2NJ}} e^{i\underline{k}_1 \cdot \underline{R}_J} \left[J^+_J, P^\alpha_J \right] b_{k_1'}$$

$$= P^\alpha_J b_{k_1} b_{k_1'} + \frac{1}{\sqrt{2NJ}} e^{i\underline{k}_1' \underline{R}_J} \left[J^+_J, P^\alpha_J \right] b_{k_i}$$

$$+ \frac{1}{\sqrt{2NJ}} e^{i\underline{k}_1 \cdot \underline{R}_J} \left[J^+_J, P^\alpha_j \right] b_{k_1'} + \frac{1}{2NJ} e^{i(\underline{k}_1 + \underline{k}_1') \underline{R}_J}$$

$$\left[J^+_J, \left[J^+_J, P^\alpha_J \right] \right] \qquad (3)$$

Proceeding on similar lines (using (2) successively)

$$(l_{k_\ell} b_{k_m} b_{k_n} b_{k_\omega} \quad l_{k_i} l_{k_\nu} l_{k_\omega}) = l_{k_\ell} l_{k_m} \quad b_{k_\mu} l_{k_\nu} P_j^\alpha b_{k_\omega}$$

in term

$$+ (l_{k_\ell} l_{k_m} \quad b_{k_\mu} l_{k_\nu}) \frac{1}{\sqrt{2NJ}} e^{i\underline{k}_\omega \underline{R}_j} [J_j^+ \; P_j^\alpha]$$

$$= l_{k_\ell} b_{k_m} \quad b_{k_\mu} P_j^\alpha b_{k_\nu} l_{k_\omega} + b_{k_\ell} b_{k_m} \quad b_{k_\mu} \frac{1}{\sqrt{NJ}} e^{i\underline{k}_\nu \underline{R}_j} [J_j, P_j^\alpha] b_{k_\omega}$$

$$+ (b_{k_\ell} l_{k_m} \quad l_{k_\mu} b_{k_\nu}) \frac{1}{\sqrt{2NJ}} e^{i\underline{k}_\mu \underline{R}_j} [J_j^+, P_j^\alpha]$$

$$= P_j^\alpha l_{k_\ell} b_{k_m} b_{k_n} \quad b_{k_\mu} b_{k_i} b_{k_\omega} + b_{k_\ell} l_{k_m} \quad l_{k_\mu} b_{k_\nu} \left(\frac{1}{\sqrt{2NJ}} e^{i\underline{k}_\ell \underline{R}_j} [J_j^+, P_j^\alpha]\right)$$

$$+ b_{k_\ell} b_{k_m} \quad b_{k_\mu} \left(\frac{1}{\sqrt{2NJ}} e^{i\underline{k}_\nu \underline{P}_j} [J_j^+ r_j^\alpha] b_{k_\omega}\right.$$

$$+ b_{k_\ell} l_{k_m} \quad \left(\frac{1}{\sqrt{2NJ}} e^{i\underline{k}_i \underline{R}_j} [J_j^+ P_j^\alpha]\right) b_{k_\mu} l_{k_\nu} l_{k_\omega} \qquad (4)$$

$$+ \frac{1}{\sqrt{2NJ}} e^{i\underline{k}_\ell \underline{R}_j} [J_j^+, P_j^\alpha] b_{k_m} b_{k_n} \quad b_{k_i},$$

Now,

$$b_{k_\ell} b_{k_m} \quad b_{k_\mu} b_{k_\nu} \left(\frac{1}{\sqrt{2NJ}} e^{i\underline{k}_\omega \underline{R}_j} [J_j^+, P_j^\alpha]\right)$$

$$= b_{k_\ell} b_{k_m} \quad l_{k_\mu} b_{k_\omega} \left(\frac{1}{\sqrt{2NJ}} e^{i\underline{k}_\omega \underline{R}_j} [J_j^+ P_j^\alpha]\right) l_{k_\ell}$$

$$+ b_{k_\ell} b_{k_m} \quad b_{k_\mu} \frac{1}{2NJ} e^{i(\underline{k}_\omega + \underline{k}_\nu) \underline{R}_j} [J_j^+, [J_j^+, P_j^\alpha]] \qquad (5)$$

$$= l_{k_2} b_{k_m} \quad b_{k_t} \left(\frac{1}{\sqrt{2NJ}} e^{i k_\omega \underline{R}_j} [J_j^+, P_j^\alpha] \right) b_{k_u} l_{k_v}$$

$$+ l_{k_1} l_{k_m} \quad b_{k_t} \left\{ \frac{1}{2NJ} e^{i(k_\omega + k_u) \underline{R}_j} [J_j^+ [J_j^+, P_j^\alpha]] \right\} b_{k_v}$$

$$+ b_{k_1} b_{k_m} \quad l_{k_u} \left\{ \frac{1}{2NJ} e^{i(k_\omega + k_v) \underline{R}_j} [J_j^+ P_j^\alpha] \right\}$$

Finally,

$$= \frac{1}{\sqrt{2NJ}} e^{i k_\omega \underline{R}_j} [J_j^+, P_j^\alpha] \, b_{k_1} b_{k_m} \quad b_{k_t} b_{k_2} l_{k_2}$$

$$+ b_{k_2} l_{k_m} \quad b_{k_u} \left\{ \frac{1}{2NJ} e^{i(k_\omega + k_v) \underline{R}_j} [J_j^+, [J_j^+ P_j^\alpha]] \right\}$$

$$+ b_{k_2} b_{k_m} \quad b_{k_t} \left\{ \frac{1}{2NJ} e^{i(k_\omega + k_u) \underline{R}_j} [J_j^+, [J_j, P_j^\alpha]] \right\} b_{k_v}$$

$$+ b_{k_2} b_{k_m} \quad b_{k_s} \left\{ \frac{1}{2NJ} e^{i(k_\omega + k_t) \underline{R}_j} [J_j^+, [J_j^+, P_j^\alpha]] \right\} l_{k_u} b_{k_v}$$

$$+ \left\{ \frac{1}{2NJ} e^{i(k_v + k_t) \underline{R}_j} [J_j^+, [J_j^+, P_j^\alpha]] \right\} b_{k_m} b_{k_n} \quad l_{k_u} l_{k_v} \qquad (6)$$

$$= \frac{1}{\sqrt{2NJ}} e^{i k_\omega \underline{R}_j} [J_j^+ P_j^\alpha] \, b_{k_2} b_{k_m} \quad b_{k_t} l_{k_u} l_{k_v}$$

$$+ \frac{e^{i k_v \underline{k}_j}}{2NJ} [J_j^+, [J_j^+, P_j^\alpha]] \sum_{l,m} e^{i k_l \underline{R}_j} \prod_{l' \neq l} l_{k_{l'}}$$

$$+ O\left(\frac{1}{N^{3/2}}\right)$$

Hence,

$$(4) = P_j^\alpha \, b_{k_l} l_{k_m} \quad b_{k_u} b_{k_v} b_{k_\omega}$$

$$+\frac{1}{\sqrt{2NJ}}\left[J_j^+, P_j^\alpha\right]\sum_{l,m}\omega\, e^{i\underline{k}_l\underline{R}_j}\prod_{l'\neq l} b_{k_{l'}}$$

$$+\frac{1}{2NJ}\,\frac{\left[J_j^+,\left[J^+ P_j^\alpha\right]\right]}{2}\sum_{l'\neq l}\sum_{l',m} e^{i\underline{k}_{l'}\underline{R}_j}\prod_{\substack{l''\neq l'\\ \neq l}} b_{k_{l''}} \qquad (6)$$

$+ O\left(\frac{1}{N^{3/2}}\right)$ whether various l are equal or not It follows from general result of (6) that

$$\prod_{l=1}^{N}(b_{k_l})^{n_{k_l}} P_j^\alpha = P_j^\alpha \prod_{l=1}^{N}(b_{k_l})^{n_{k_l}}$$

$$+\frac{1}{\sqrt{2NJ}}\left[J_j^+ P_j^\alpha\right]\sum_l n_{k_l} e^{i\underline{k}_l\underline{R}_j}(b_{k_l})^{n_{k_l}-1}\prod_{l'\neq l}(b_{k_{l'}})^{n_{k_{l'}}}$$

$$+\frac{1}{2NJ}\,\frac{\left[J_j^+,\left[J_j^+, P_j^\alpha\right]\right]}{2}\sum_l n_{k_l}(n_{k_l}-1)\, e^{2i\underline{k}_l\underline{R}_j}(b_{k_l})^{n_{k_l}-2}\prod_{l\neq l'}(b_{k_{l'}})^{n_{k_{l'}}}$$

$$+\frac{1}{2NJ}\,\frac{\left[J_j^+,\left[J_j^+, P_j^\alpha\right]\right]}{2}\sum_l n_{k_l}\, e^{2i\underline{k}_l\underline{R}_j}(b_{k_l})^{n_{k_l}-1}\sum_{l'\neq l} n_{k_{l'}}(b_{k_{l'}})^{n_{k_{l'}}-1}\prod_{l'\neq l}(b_{k_{l'}})^{n_{k_{l'}}}$$

$$+\frac{1}{2NJ}\,\frac{\left[J_j^+,\left[J_j^+, P_j^\alpha\right]\right]}{2}\sum_{l\neq l'} n_{k_l} n_{k_{l'}}\, e^{i\underline{k}_l\underline{R}_j} e^{i\underline{k}_{l'}\underline{R}_j}(b_{k_l})^{n_{k_l}-1}(b_{k_{l'}})^{n_{k_{l'}}-1}\prod_{l''\neq l,l'}(b_{k_{l''}})^{n_{k_{l''}}}$$

$$+ O\left(\frac{1}{N^{3/2}}\right) \qquad (7)$$

To evaluate $\langle a|P_j^\alpha|a'\rangle$

where $|a\rangle = |n_{k_1}, n_{k_2} \ldots n_{k_N}\rangle$, $\sum_1^N n_{k_l} = n$

$|a'\rangle = |n'_{k_1}, n'_{k_2}, \ldots n'_{k_N}\rangle$, $\sum_1^N n'_{k_l} = n'$

So,

$$\langle a|P_J^\alpha|a'\rangle$$

$$= \langle 0| \prod_{\iota=1}^{N} \frac{(b_{k_\iota})^{n_{k_\iota}}}{\sqrt{n_{k_\iota}!}} P_J^\alpha \prod_{\iota=1}^{N} \frac{(b_{k_\iota}^+)^{n_{k_\iota}}}{\sqrt{n_{k_\iota}!}} |0\rangle$$

$$= \langle 0| P_J^\alpha \prod_{\iota=1}^{N} \frac{(b_{k_\iota})^{n_{k_\iota}}}{\sqrt{n_{k_\iota}!}} \prod_{\iota=1}^{N} \frac{(b_{k_\iota}^+)^{n_{k_\iota}}}{\sqrt{n_{k_\iota}!}} |0\rangle$$

$$+ \frac{1}{\sqrt{2NJ}} \langle 0| [J_J^+ P_J^\alpha] \sum_\iota \frac{n_{k_\iota}}{\sqrt{n_{k_\iota}!}} e^{i\underline{k}_\iota \underline{R}_J} (b_{k_\iota})^{n_{k_\iota}-1} \prod_{\iota' \neq \iota} \frac{(b_{k_{\iota'}})^{n_{k_{\iota'}}}}{\sqrt{n_{k_{\iota'}}!}} \prod_{\iota} \frac{(b_{k_\iota}^+)^{n_{k_\iota}}}{\sqrt{n_{k_\iota}!}} |0\rangle$$

$$+ \frac{1}{2NJ} \langle 0| \frac{[J_J^+, [J_J^+ P_J^\alpha]]}{2} \sum_\iota \frac{n_{k_\iota}}{\sqrt{n_{k_\iota}}} e^{i\underline{k}_\iota \underline{R}_J} (b_{k_\iota})^{n_{k_\iota}-1} \sum_{\iota' \neq \iota} \frac{n_{k_{\iota'}}}{\sqrt{n_{k_{\iota'}}!}} e^{i\underline{k}_{\iota'} \underline{R}_J} (b_{k_{\iota'}})^{n_{k_{\iota'}}-1}$$

$$\times \prod_{\substack{\iota'' \neq \iota \\ \neq \iota'}} \frac{(b_{k_{\iota''}})^{n_{k_{\iota''}}}}{\sqrt{n_{k_{\iota''}}!}} \prod_{\iota} \frac{(b_{k_\iota}^+)^{n_{k_\iota}}}{\sqrt{n_{k_\iota}!}} |0\rangle + O\left(\frac{1}{N^{3/2}}\right) \qquad (8)$$

<u>Case I</u> $n = n'$ If $n_{k_1} = n'_{k_1}$ for $\iota = 1$

$$\langle a|P_J^\alpha|a'\rangle = \langle 0|P_J^\omega|0\rangle$$

$$+ \frac{1}{\sqrt{2NJ}} \langle 0| [J_J^+, P_J^\alpha] \sum_\iota n_{k_\iota} e^{i\underline{k}_\iota \underline{R}} |1_{k_\iota}\rangle$$

$$+ \frac{1}{2NJ} \langle 0| \frac{[J_J^+, [J_J^+, P_J^\alpha]]}{2} \sum_\iota n_{k_\iota} \sum_{\iota' \neq \iota} n_{k_{\iota'}} e^{i\underline{k}_\iota \underline{R}_J + i\underline{k}_{\iota'} \underline{R}_J} |1_{k_{\iota'}}, 1_{k_\iota}\rangle \qquad (9)$$

$$+ O\left(\frac{1}{NJ^{3/2}}\right)$$

$$= \langle 0|P_J^\alpha|0\rangle$$

$$+ \left(\frac{1}{\sqrt{2NJ}}\right)^2 \sum_l n_{k_l} \langle 0|[J_J^+, P_J^\alpha] J_J^-|0\rangle$$

$$+ O\left(\frac{1}{N^2}\right) \qquad (10)$$

If $n_{k_l} = n'_{k_l}$ except $n_{k_{l'}} = n'_{k_{l'}} + r$

and $n_{k_{l''}} = n'_{k_{l''}} - r$

So that $n = n'$, then for $r = 1$,

$$\langle a|P_J^\alpha|a'\rangle = \langle n_{k_l}, \; n'_{k_{l'+1}}, \; n'_{k_{l'}} - 1, \; n_{k_N}|P_J^\alpha|n'_{k_l} \; n'_{k_{l'}}, n'_{k_{l''}} \; n_{k_N}\rangle$$

$$= \left(\frac{1}{\sqrt{2NJ}}\right)^2 \langle 0|[J_J^+ P_J^\alpha]\left[\left\{\sum_{\substack{l\neq l' \\ \neq l''}} \frac{n_{k_l}}{\sqrt{n_{k_l}}} e^{i\underline{k}_l \cdot \underline{R}_J} J_J^-\right\}\right.$$

$$\left. + n_{k_{l'}} e^{i\underline{k}_{l'} \cdot \underline{R}_J} + \frac{n_{k_{l''}}}{\sqrt{2}} e^{i\underline{k}_{l'} \cdot \underline{R}_J - 2\,\underline{k}_{l''} \cdot \underline{R}_J} J_J^- J_J^-\right]|0\rangle$$

$$+ O\left(\frac{1}{N^{3/2}}\right) \qquad (11)$$

For $r \geq 2$, (8) yields zero upto order of $\left(\frac{1}{N^{3/2}}\right)$

Case II $\quad n = n' + 1$, let $n_{k_1} = n'_{k_1}$ except $n_{k_{1'}} = n'_{k_{1'}} + 1$

$$\langle a|P^{\alpha}_{J}|a'\rangle$$

$$= \frac{1}{\sqrt{2NJ}} \langle 0| \left[J^{+}_{J}, P^{\alpha}_{J} \right] \sqrt{n_{k_{1'}}}\; e^{i \underline{k}_{1'} \cdot \underline{R}_{J}} |0\rangle$$

$$+ O\left(\frac{1}{N^{3/2}} \right) , \text{ the first term of (8) being zero} \qquad (12)$$

and for all other distributions of m gnons, it is only $O\left(\frac{1}{N^{3/2}} \right)$, hence negligible

Case III

$n = n' - 1$, let $n_{k_1} = n'_{k_1}$ except $n_{k_{1'}} = n'_{k_{1'}} - 1$

First term of (8) will contribute to $\langle a|P^{\alpha}_{J}|a'\rangle$

$$= \frac{1}{\sqrt{2NJ}}\; e^{-i\underline{k}_{1'} \cdot \underline{R}_{J}} \langle 0|P^{\alpha}_{J} J^{-}_{J}|0\rangle \sqrt{n_{k_{1'}} + 1}$$

$$+ O\left(\frac{1}{N^{3/2}} \right) \qquad (13)$$

Case IV $\quad |n-n'| > 2 = 1$ say

Then

$$\langle a'|P^{\alpha}_{J}|a\rangle \sim \frac{1}{N^{1/2}}$$

and contribution of such terms to scattering cross section would be of the order of $\frac{1}{N^{1/2}}$ i e., at the most $\frac{1}{N^{2}}$ Hence safely neglected for one magnon processes

Hence, using (10), (11), (12) and (13) we get,

$$\langle P_j^{\alpha}(0)\, P_{j'}^{\beta}(t)\rangle$$

$$= \sum_{a,a'} e^{-E_a\beta'} \langle a|P_j^{\alpha}|a'\rangle\langle a'|P_{j'}^{\beta}|a\rangle \frac{\exp\{\frac{it}{\hbar}(E_a - F)\}}{\sum_a e^{-E_c\beta'}}$$

$$= \Bigg[\sum_{\{n_{k_i}\}} e^{\sum_i n_{k_i}\epsilon_{k_i}\beta'} \Big\{\delta\langle 0|P_j^{\alpha}|0\rangle + \frac{1}{2NJ}\sum_i n_{k_i}\langle 0|[J_j^{+}, P_j^{\alpha}]J_j^{-}|0\rangle\Big\}$$

$$\times\Big\{\langle 0|P_{j'}^{\beta}|0\rangle + \frac{1}{2NJ}\sum_i n_{k_i}\langle 0|[J_j^{+}P_{j'}^{\beta}]J_{j'}^{-}|0\rangle\Big\}$$

$$+\sum_i \frac{\sqrt{n_{k_i}}}{2NJ}\langle 0|[J_j^{+}, P_j^{\alpha}]|0\rangle\, e^{i\underline{k}_i\underline{R}_j}\sqrt{n_{k_i}}\langle 0|P_{j'}^{\beta}J_{j'}^{-}|0\rangle\, e^{-i\underline{k}_i\underline{R}_j} e^{-\frac{it}{\hbar}\epsilon_{k_i}}$$

$$+\sum_i \frac{\sqrt{n_{k_i}+1}}{2NJ}\langle 0|P_j^{\alpha}J_j^{-}|0\rangle\, e^{-i\underline{k}_i\underline{R}_j}\sqrt{n_{k_i}+1}\langle 0|[J_j^{+}, P_{j'}^{\beta}]|0\rangle\, e^{i\underline{k}_i\underline{R}_j} e^{\frac{it}{\hbar}\epsilon_k}\Big\}\Bigg]\times$$

$$\times\frac{1}{\sum_{\{n_{k_i}\}} e^{-\sum_i \epsilon_{k_i} n_{k_i}\beta'}} + O\left(\frac{1}{N^2}\right) \qquad (14)$$

It is to be noted that we have not made use of the explicit form of $|0\rangle$ to obtain (14) The only assumption is that $|0\rangle$ is product of individual ionic wavefunction by virtue of ions being independent But the conversion of magnon operators to ionic operators has been done for a ferromagnet

Also for ferromagnet,

$$|0\rangle = |JJ\rangle_1 \, |JJ\rangle_2 \quad \cdots \quad |JJ\rangle_N \tag{15}$$

So, (14) becomes

$$\begin{aligned}
&\langle JJ|P_j^{\alpha}|JJ\rangle_j \langle JJ|P_{j'}^{\beta}|JJ\rangle_{j'} + \frac{1}{2NJ}\sum_i \langle n_{k_i}\rangle \Big\{ \langle JJ|[J_j^{+} P_j^{\alpha}]J_1^{-}|JJ\rangle_j \\
&\times \langle JJ|P_j^{\beta}|JJ\rangle_{j'} + \langle JJ|P_j^{\alpha}|JJ\rangle_j \langle JJ|[J_{j'}^{+} P_j^{\beta}]J_j^{-}|JJ\rangle_{j'} \Big\} \\
&+ \sum_i \frac{\langle n_{k_i}\rangle}{2NJ} \langle JJ|[J_j^{+}, P_j^{\alpha}]|JJ\rangle_j \langle JJ|P_{j'}^{\beta} J_{j'}^{-}|0\rangle e^{i\underline{k}_i(\underline{R}_j - \underline{R}_{j'})} e^{-\frac{it}{\hbar}\epsilon_{k_i}} \\
&+ \sum_i \frac{\langle n_k\rangle + 1}{2NJ} \langle JJ|P_j^{\alpha} J_j^{-}|JJ\rangle_j \langle JJ|[J_{j'}^{+}, P_{j'}^{\beta}]|JJ\rangle_{j'} e^{-i\underline{k}_i(\underline{R}_j - \underline{R}_{j'})} e^{\frac{it}{\hbar}\epsilon_k} \\
&+ O\left(\frac{1}{N^2}\right)
\end{aligned} \tag{16}$$

$$\begin{aligned}
&= \langle JJ|P_j^{\alpha}|JJ\rangle_j \langle JJ|P_{j'}^{\beta}|JJ\rangle_{j'} + \frac{1}{N}\sum_i \langle n_{k_i}\rangle \Big[\Big\{ \langle JJ|P_j^{\alpha}|JJ\rangle_j - \langle JJ|P_j^{\alpha}|JJ\rangle_j \Big\} \\
&\times \langle JJ|P_{j'}^{\beta}|JJ\rangle_{j'} + \langle JJ|P_j^{\alpha}|JJ\rangle_j \Big\{ \langle JJ|P_{j'}^{\beta}|JJ\rangle_{j'} - \langle JJ|P_{j'}^{\beta}|JJ\rangle_{j'} \Big\} \Big] \\
&+ \sum_i \frac{\langle n_{k_i}\rangle}{N} \langle JJ|P_j^{\alpha}|JJ\rangle_j \langle JJ|P_{j'}^{\beta}|JJ\rangle_{j'} e^{i\underline{k}_i(\underline{R}_j - \underline{R}_{j'})} e^{-\frac{it}{\hbar}\epsilon_k} \\
&+ \sum_i \frac{\langle n_{k_i}\rangle + 1}{N} \langle JJ|P_j^{\alpha}|JJ\rangle_j \langle JJ|P_{j'}^{\beta}|JJ\rangle_{j'} e^{-i\underline{k}_i(\underline{R}_j - \underline{R}_{j'})} e^{\frac{it}{\hbar}\epsilon_k}
\end{aligned} \tag{17}$$

From (17) we can immediately obtain the differential scattering cross sections

For spin only case replace $|JM\rangle$ by $|SM_s\rangle$ and $\bar{P}_J$ by $F(q)\bar{S}_J$ then (17) reduces correctly to that for spin only case

CHAPTER - IV

SCATTERING CROSS-SECTION FOR ANTIFERROMAGNETIC RARE EARTH CRYSTALS

4 1 ANTIFERROMAGNETIC GROUND STATE AS ORDERED STATE

In this study, ordered state is taken as ground state Details of antiferromagnetic magnons are given in Kittel, Q Th S (page 58) and for ground state J M Ziman

$$H = -N(We + V_1) + \sum_k W_k(\alpha_k^+\alpha_k + \beta_k^+\beta_k + 1) \tag{18}$$

$$\text{with } \alpha_k = U_k C_k - v_k d_k^+ , \ \beta_k = U_k d_k - v_k C_k^+ \tag{19}$$

$$\alpha_k^+ = U_k C_k^+ - v_k d_k , \ \beta_k^+ = U_k d_k^+ - v_k C_k$$

$$\text{and, } C_k = \frac{1}{\sqrt{N}} \sum_j e^{i\underline{k}\,\underline{R}_j} a_j , \ d_k = \frac{1}{\sqrt{N}} \sum_l e^{i\underline{k}\,\underline{R}_l} b_l \tag{20}$$

each sublattice has N atoms

$$Ja_j^+ = \sqrt{2J}\ a_j , \ Ja_j^- = \sqrt{2J}\ a_j^+ , \ Ja_j^z = J - a_j^+a_j \tag{21}$$

$$J_{b_l}^+ = \sqrt{2J}\ b_l^+ , \ J_{b_l}^- = \sqrt{2J}\ b_l , \ J_{b_l}^z = -J + b_l^+b_l$$

so that a_j's and b_j's satisfy the same commutation relations, i e , for Bosons In order that α_k , β_k be bose operators and Hamiltonian be diagonal, we must have

$$U_k^2 - v_k^2 = 1 \tag{22}$$

$$r_k(U_k^2 + v_k^2) + (1 + 2/\mu_o H_1)\ 2\ U_k v_k = 0$$

Moreover, the assumption implicit in writing the Bo e operators (i e , the spin wave assumption for antiferromagnetic) is that $\langle a_j^+ a_j \rangle/2S$ and $\langle b_l^+ b_l \rangle/2S$ are much smaller than unity (see exact Holstein Primakoff transf) Hence, it amounts to taking $J^z a_j$ and $J^z b_l$ close to J and -J respectively Hence under this assumption, the ground state of antiferr is $\uparrow\downarrow\uparrow\downarrow\uparrow\downarrow\uparrow\downarrow$ But this configuration is only a approx to ground tate and not the true ground state The best proof of existence of two sublattice configuration with opposite orientation is given by neutron diffraction experiments

4.2 CALCULATION OF MATRIX ELEMENTS

Magnon eigen state for antiferromagnet is

$$|\lambda\rangle = |n_{k_1}\alpha,\ n_{k_2}\alpha, \quad n_{k_N}\alpha,\ n_{k_1}\beta,\ n_{k_2}\beta, \quad n_{k_N}\beta\rangle$$

$$= \prod_l \frac{(\alpha^+_{k_l})^{n_{k_l}\alpha}}{\sqrt{n_{k_l}\alpha!}} \frac{(\beta^+_{k_l})^{n_{k_l}\beta}}{\sqrt{n_{k_l}\beta!}} |0\rangle \qquad (23)$$

Now

$$\alpha_{k_l} = \frac{U_{k_l}}{\sqrt{N}} \sum_{j_1} e^{i\underline{l}_1 \underline{R}_{j1}} a_{j_1} - \frac{v_{l_1}}{\sqrt{N}} \sum_{e_1} e^{-i\underline{k}_1 \underline{R}_{e_1}} b^+_{l_1}$$

$$= \frac{U_{k_1}}{\sqrt{N\ 2J}} \sum_{j_1 \neq j} e^{i\underline{l}'_1 \underline{R}_{j_1}} J^+ a_{j_1} - \frac{v_{k_1}}{\sqrt{2JN}} \sum_{l_1 \neq l} e^{-i\underline{k}_1 \underline{R}_{l_1}} J^+ b_{l_1}$$

$$+ \frac{U_{k_1}}{\sqrt{2NJ}} e^{i\underline{k}_1 \underline{R}_J} J a_J^+ - \frac{V_{k_1}}{\sqrt{2NJ}} e^{-i\underline{k}_1 \underline{R}_1} J b_1^+ \qquad (24)$$

Now,

$$\left[\alpha_{k_1}, P_{a_J}^{\alpha'} \right] = \frac{U_{k_1}}{\sqrt{2NJ}} e^{i\underline{k}_1 \underline{R}_J} \left[J a_J^+ , P a_J' \right]$$

and

$$\left[\alpha_{k_1}, P_{b_1}^{\alpha'} \right] = \frac{-V_{k_1}}{\sqrt{2NJ}} e^{-i\underline{k}_1 \underline{R}_1} \left[J b_1^+ , P_{b_1}^{\alpha'} \right] \qquad (25)$$

Now, proceeding on similar lines as for one sublattice ferromagnet, we obtain

$$\prod_{1=1}^{N} (\alpha_{k_1})^{n_{k_1}\alpha} P_{a_J}^{\alpha'}$$

$$= P_{a_J}^{\alpha'} \prod_{1=1}^{N} (\alpha_{k_1})^{n_{k_1}\alpha}$$

$$+ \frac{1}{\sqrt{2NJ}} \left[J_{a_J}^+ , P_J^{\alpha'} \right] \sum_1 U_{k_1} n_{k_1} e^{i\underline{k}_1 \underline{R}_J} \times$$

$$(\alpha_{k_1})^{n_{k_1}\alpha-1} \prod_{1' \neq 1} (\alpha_{k_{1'}})^{n_{k_{1'}}\alpha}$$

$$+ \frac{1}{2NJ} \frac{\left[J_{a_J}^+ , \left[J_{a_J}^+ , P_{a_J}^{\alpha'} \right] \right]}{2} \left\{ \sum_1 U_{k_1} n_{k_1} (\alpha_{k_1})^{n_{k_1}\alpha-1} \times \right.$$

$$e^{i\underline{k}_1 \cdot \underline{R}_J} \times \sum_{1' \neq 1} U_{k_{1'}} n_{k_{1'}}\alpha (\alpha_{k_{1'}})^{n_{k_{1'}}\alpha-1} e^{i\underline{k}_{1'} \underline{R}_J}$$

$$\prod_{\substack{1'' \neq 1 \\ \neq 1'}} (\alpha_{k_{i''}})^{n_{k_1''}\alpha}$$

$$+ \sum_{1} U_{k_1}^2 \; n_{1\,1}{}^{\alpha} \; (n_{k_1}{}^{\alpha}-1) \; e^{-2i\underline{k}_1 \underline{R}_J} \; (\alpha_{k_1})^{n_{k_1}{}^{\alpha}-2} \prod_{1' \neq 1} (\alpha_{1'})^{n_{1'}{}^{\alpha}} \Big\}$$

$$+ O\left(\frac{1}{N^{3/2}}\right) \tag{26}$$

Again,

$$\prod_{1=1}^{N} (\alpha_{k_1})^{n_{k_1}{}^{\alpha}} \; P_{b_1}^{\alpha'} = P_{b_1}^{\alpha'} \prod_{1=1}^{N} (\alpha_{k_1})^{n_{k_1}{}^{\alpha}} + \frac{1}{\sqrt{2NJ}} \left[J_{b_1}^{+}, P_{b_1}^{\alpha'}\right] \times$$

$$\sum_{1} (-v_{k_1})^{n_{k_1}{}^{\alpha}} \; (\alpha_{k_1})^{n_{k_1}{}^{\alpha-1}} \; e^{-i\underline{k}_1 \underline{I}_1}$$

$$\prod_{1' \neq 1} (\alpha_{k_1'})^{n_{k_1'}{}^{\alpha}} + \frac{1}{2NJ} \; \frac{\left[J_{b_1}^{+}, \left[J_{b_1}^{+}, P_{b_1}^{\alpha'}\right] I\right]}{2} \times$$

$$\Big\{ \sum_{1} v_{k_1} \; n_{k_1}{}^{\alpha} \; (\alpha_{k_1})^{n_{k_1}{}^{\alpha-1}} \; e^{-i\underline{k}_1 \underline{R}_1} \; \gamma \sum_{1' \neq 1} v_{k_1'} \; n_{1\,1'}{}^{\alpha} \times$$

$$(\alpha_{k_1'})^{n_{k_1'}{}^{\alpha-1}} \; e^{-i\underline{k}_1' \underline{R}_1} \prod_{\substack{1'' \neq 1' \\ \neq 1}} (\alpha_{k_1''})^{n_{k_1''}{}^{\alpha}}$$

$$+ \sum_{1} v_{1\,1}^2 \; n_{1\,1}{}^{\alpha} \; (n_{k_1}{}^{\alpha}-1) \times e^{-2i\underline{k}_1 \underline{R}_1} \; (\alpha_{1_1})^{n_{k_1}{}^{-2}}$$

$$\prod_{1' \neq 1} (\alpha_{k_1'})^{n_{k_1'}{}^{\alpha}} + O\left(\frac{1}{N^{3/2}}\right) \tag{27}$$

Now

$$\beta_{k_1} = \frac{U_{k_1}}{\sqrt{2NJ}} \sum_{l_1 \neq l} e^{i k_{l_1} R_{l_1}} J^-_{b_{l_1}} - \frac{v_{k_1}}{\sqrt{2NJ}} \sum_{J_1 \neq J} e^{-i k_{J_1} R_{J_1}} J^-_{a_{J_1}}$$

$$+ \frac{U_{k_1}}{\sqrt{2NJ}} e^{i k_1 R_1} J^-_{b_1} - \frac{v_{k_1}}{\sqrt{2NJ}} e^{-i k_1 R_J} J^-_{a_J}$$

$$\prod_l (\beta_{k_l})^{n_{k_l}\beta} \; P^{\alpha'}_{a_J}$$

$$= P^{\alpha'}_{a_J} \prod_l (\beta_{k_l})^{n_{k_l}\beta} + \frac{1}{\sqrt{2NJ}} \left[J^-_{a_J}, P^{\alpha'}_J \right] \sum_l (-v_{k_l}) n_{k_l} (\beta_{k_l})^{n_{k_l}\beta - 1}$$

$$e^{-i k_1 R_J} \prod_{l' \neq i} (\beta_{k_{i'}})^{n_{k_{l'}}\beta} + \frac{1}{2NJ} \frac{\left[J^-_{a_J}, \left[J^-_{a_J}, P^{\alpha'}_{a_J} \right] \right]}{2} \times$$

$$\left\{ \sum_l v_{k_l} n_{k_l}\beta \, (\beta_{k_l})^{n_{k_l}\beta - 1} \; e^{-i k_1 R_J} \sum_{l' \neq l} v_{l'} \, n_{k_{l'}}\beta \right.$$

$$\times \, (\beta_{k_{l'}})^{n_{k_{l'}}\beta - 1} \; e^{-i k_1 R_J} \prod_{\substack{l'' \neq l \\ \neq l'}} (\beta_{k_{l''}})^{n_{l''}\beta}$$

$$\left. + \sum_l v^2_{k_i} \, n_{k_l}\beta (n_{k_l}\beta - 1) \; e^{-2 i k_1 R_J} (\beta_l)^{n_{k_l}\beta - 2} \prod_{l' \neq l} (\beta_{k_{l'}})^{n_{k_{l'}}\beta} \right.$$

$$+ O\left(\frac{1}{N^{3/2}} \right) \qquad (28)$$

and,

$$\prod_{l} (\beta_{k_l})^{n_{k_l}\beta} \; P^{\alpha'}_{b_l}$$

$$= P^{\alpha'}_{b_l} \prod_{l} (\beta_{k_l})^{n_{k_l}} + \frac{1}{\sqrt{2NJ}} \left[J^{-}_{b_l}, P^{\alpha'}_{b_l} \right] \sum_{l} U_{k_l} n_{l\,l} (\beta_{l\,l})^{n_{k_l}\beta - 1}$$

$$e^{+il\underline{k}_l \underline{R}_l} \times \prod_{l' \neq l} (\beta_{l\,l'})^{n_{k_{l'}}\beta} + \frac{1}{2NJ} \frac{\left[J^{-}_{b_l}, \left[J^{-}_{b_l}, P^{\alpha'}_{b_l} \right] \right]}{2} \times$$

$$\left\{ \sum_{l} U_{k_l} n_{k_l\beta} (\beta_{k_l})^{n_{k_l}\beta - 1} e^{il\underline{k}_l \underline{R}_l} \times \sum_{l' \neq l} U_{l\,l'} n_{l\,l'\beta} (\beta_{k_{l'}})^{n_{k_{l'}}\beta - 1} \right.$$

$$e^{il\underline{k}_{l'} \underline{R}_l} \prod_{\substack{l'' \neq l' \\ \neq l}} (\beta_{l\,l'})^{n_{k_{l'}}\beta} + \sum_{l} U^2_{k_l} n_{l\,l\alpha} (n_{l\,l} - 1)$$

$$\left. e^{2il\underline{k}_l \underline{R}_l} (\beta_{k_l})^{n_{l\,l}\alpha - 2} \prod_{l' \neq l} (\beta_{k_{l'}})^{n_{k_{l'}}\alpha} \right\} + O\left(\frac{1}{N^{3/2}} \right)$$

(29)

We want to evaluate $\langle A | P^{\alpha}_{a_j} | A' \rangle$, $\langle A | P^{\alpha}_{b_l} | A' \rangle$ etc

Number of magnons in $|A\rangle$ is $n\alpha + nJ$

Number of magnons in $|A'\rangle$ is $n'\alpha + n'\alpha$

$$\langle A | P^{\alpha}_{a_j} | A' \rangle$$

$$= \langle 0 | \left\{ \prod_{l} \frac{(\alpha_{k_l})^{n_{k_l}\alpha}}{\sqrt{n_{k_l\alpha}!}} \frac{(\beta_{k_l})^{n_{k_l}\beta}}{\sqrt{n_{k_l\beta}!}} \right\} P^{\alpha}_{a_j} \left\{ \prod_{l} \frac{(\alpha^{+}_{k_l})^{n_{k_l}\alpha}}{\sqrt{n_{k_l\alpha}!}} \frac{(\beta^{+}_{k_l})^{n_{k_l}\beta}}{\sqrt{n_{k_l\beta}!}} \right\} | 0 \rangle$$

(30)

Now

$$\prod_{l}(\alpha_{k_l})^{n_{k_l\alpha}}\,(\beta_{k_l})^{n_{k_l\beta}}\,P^{\alpha'}_{a_J}$$

$$= \prod_{l}(\alpha_{k_l})^{n_{k_l\alpha}}\,P^{\alpha'}_{a_J}\prod_{l}(\beta_{l_l})^{n_{k_l\beta}} + \prod_{l}(\alpha_{l_l})^{n_{l_l\alpha}}\times$$

$$\frac{1}{\sqrt{2NJ}}\left[J^{-}_{a_J},P^{\alpha'}_{J}\right]\sum_{l}(-v_{k_l})\,n_{l_l\beta}\,(\beta_{k_l})^{n_{l_l\beta-1}}\,e^{-il\underline{k}_l\cdot\underline{R}_J}\times$$

$$\prod_{l'\neq l}(\beta_{k_{l'}})^{n_{k_{l'}\beta}} + \frac{1}{2NJ}\prod_{l}(\alpha_{k_l})^{n_{k_l\alpha}}\,\frac{\left[J^{-}_{a_J},\left[J^{-}_{a_J},P^{\alpha'}_{a_J}\right]\right]}{2}\times$$

$$\left\{\sum_{l}v_{k_l}\,n_{k_l\beta}(\beta_{k_l})^{n_{k_l\beta-1}}\,e^{-il\underline{k}_l\cdot\underline{R}_J}\times\sum_{l'\neq l}v_{k_{l'}}\,n_{l_{l'}\beta}\right.$$

$$(\beta_{k_{l'}})^{n_{k_{l'}\beta-1}}\times e^{-il\underline{k}_l\cdot\underline{R}_J}\,e^{-il\underline{k}_{l'}\cdot\underline{R}_J}\prod_{l'\neq l}(\beta_{l_{l'}})^{n_{l_{l'}\beta}}$$

$$\left.+\sum_{l}v^2_{k_l}\,n_{k_l\beta}(n_{k_l\beta-1})\,e^{-2il\underline{k}_l\cdot\underline{R}_J}\,(\beta_{k_l})^{n_{l_{l'}\beta-2}}\prod_{l'\neq l}(\beta_{k_{l'}})^{n_{k_{l'}\beta}}\right\}$$

$$+\;O\left(\frac{1}{N^{3/2}}\right) \qquad (31)$$

$$= P^{\alpha'}_{a_J}\prod_{l}(\alpha_{k_l})^{n_{k_l\alpha}}\,(\beta_{k_l})^{n_{k_l\beta}} + \frac{1}{\sqrt{2NJ}}\left[J^{+}_{a_J},P^{\alpha'}_{J}\right]\sum_{l}U_{k_l}\,n_{l_l\alpha}\times$$

$$e^{i\underline{k}_1 \underline{R}_J} (\alpha_{k_1})^{n_{k_1}\alpha-1} \prod_{1' \neq 1} (\alpha_{k_{1'}})^{n_{k_{1'}}\alpha} \times \prod_{1_1} (\quad_{1_1})^{n_{k_{1_1}}C} + \frac{1}{2NJ} >$$

$$\frac{[J^+_{a_J}, [J^+_{a_J}, P^{\alpha'}_J]]}{2} \Big\{ \sum_1 U_{1\,1}\, n_{k_1\alpha} (\alpha_{1\,1})^{n_{k_1}\alpha-1} e^{i\underline{l}_{-1}\underline{R}_J}$$

$$\times \sum_{1' \neq 1} U_{1\,1'}\, n_{k_{1'}\alpha} (\alpha_{k_{1'}})^{n_{k_{1'}}\alpha-1} e^{i\underline{k}_{1'}\underline{R}_J} \prod_{\substack{1'' \neq 1 \\ \neq J'}} (\alpha_{k_{1''}})^{n_{k_{1''}}\alpha} \Big\}$$

$$+ \sum_1 U^2_{k_1}\, n_{k_1\alpha}(n_{k_1\alpha}-1)\, e^{2i\underline{k}_1 \underline{R}_J} (\alpha_{k_1})^{n_{k_1}\alpha-2} \prod_{1' \neq 1} (\alpha_{1\,1'})^{n_{k_{1'}}\alpha} \Big\}$$

$$\prod_{1_1} (\beta_{k_{1_1}})^{n_{k_{1_1}}\beta}$$

$$+ \frac{1}{\sqrt{2NJ}} \left[J^-_{a_J}, P^{\alpha'}_J\right] \sum_1 (-v_{k_1})\, n_{1\,1\beta} (\beta_{k_1})^{n_{k_1}\beta-1} e^{-i\underline{l}_{-1}\underline{R}_J} \times$$

$$\prod_{1' \neq 1} (\beta_{k_{1'}})^{n_{k_{1'}}\beta} \prod_{1_1} (\alpha_{k_{1_1}})^{n_{k_{1_1}}\alpha} + \frac{1}{2NJ} \left[J^-_{a_J}, [J^-_{a_J}, P^{\alpha'}_J]\right] \times$$

$$\sum_1 U_{k_1}\, n_{k_1\alpha}\, e^{i\underline{k}_1 \underline{R}_J} (\alpha_{k_1})^{n_{k_1}\alpha-1} \prod_{1' \neq 1} (\alpha_{1\,1'})^{n_{1\,1'}\alpha}$$

$$\times \sum_{1_1} (-v_{k_{1_1}})\, n_{k_{1_1}\beta} (\beta_{k_{1_1}})^{n_{k_{1_1}}\beta-1} e^{-i\underline{k}_1 \underline{R}_J} \prod_{1'_1 \neq 1_1} (\quad_{k_{1'_1}})^{n_{k_{1'_1}}\beta}$$

$$+ \frac{1}{2NJ} \frac{\left[J^-_{a_J}, \left[J^-_{a_J}, P^{\alpha'}_{a_J} \right] \right]}{2} \Bigg\{ \sum_{l} v_{k_l} n_{k_l} \left(\quad \right)^{n_{k_l}\gamma - 1}$$

$$e^{-i\underline{k}_l \underline{R}_J} \sum_{l' \neq l} v_{k_{l'}} n_{k_{l'}\gamma} \left(\quad \right)^{n_{k_{l'}}\rho - 1} e^{-i\underline{k}_{l'} \underline{R}_J}$$

$$\prod_{\substack{l'' \neq l \\ \neq l'}} \left(\quad \right)^{n_{k_{l''}}\beta} \prod_{l_1} \left(\alpha_{k_{l_1}} \right)^{n_{k_{l_1}}\alpha} + \sum_{l} v^2_{k_l} n_{k_l} \left(\quad \right) e^{-2i\underline{k}_l \underline{R}_J}$$

$$\times \left(\beta_{k_l} \right)^{n_{k_l}\gamma - 2} \prod_{l' \neq l} \left(\beta_{k_{l'}} \right)^{n_{k_{l'}}} \prod_{l_1} \left(\alpha_{k_{l_1}} \right)^{n_{k_{l_1}}\alpha} \Bigg\} + O\left(\frac{1}{N^{3/2}} \right)$$

(32)

Case I $\quad n_{k_l\alpha} = n'_{k_l\alpha}$ and $n_{k_l\gamma} = n'_{k_l\gamma}$,

$$\langle A | P^{\alpha'}_{a_J} | A' \rangle$$

$$= \langle 0 | P^{\alpha'}_{a_J} | 0 \rangle + \frac{1}{\sqrt{2NJ}} \langle 0 | \left[J^+_{a_J}, P^{\alpha'}_{J} \right] \sum_{l} U_{l} \, n_{k_l\alpha} \, e^{i\underline{k}_l \underline{R}_J} | 1_{k_l\alpha} \rangle$$

$$+ \frac{1}{\sqrt{2NJ}} \langle 0 | \left[J^-_{a_J}, P^{\alpha'}_{J} \right] \sum_{l} (-v_{k_l}) n_{k_l\gamma} | 1_{k_l\gamma} \rangle e^{-i\underline{k}_l \underline{R}_J} + O\left(\frac{1}{N^2} \right)$$

$$= \langle 0 | P^{\alpha'}_{a_J} | 0 \rangle + \frac{1}{2NJ} \langle 0 | \left[J^+_{a_J}, P^{\alpha'}_{J} \right] J^-_{a_J} | 0 \rangle \sum_{l} U^2_{k_l} \, n_{k_l\alpha} + O\left(\frac{1}{N^2} \right)$$

(33)

Again,

$$\langle A|P^{\alpha'}_{b_1}|A'\rangle$$

$$= \langle 0|P^{\alpha}_{b_1}|0\rangle + \frac{1}{2NJ}\langle 0|\left[J^{-}_{b_1}, P^{\alpha'}_{b_1}\right] J^{+}_{b_1}|0\rangle \sum_{k_1} U^2_{k_1} n_{k_1 r} + O\left(\frac{1}{N^2}\right)$$

Case II

It is easy to see that matrix element $\langle A|P^{c'}_{a_J}|A'\rangle$ for which $n_\alpha + n_\beta = n'_\alpha + n'_\beta$ but $n_{k_1\alpha} \neq n'_{k_1\alpha}$ and $n_{k_1\beta} \neq n'_{k_1\beta}$,

$$\langle A | P^{\alpha'}_{a_J} | A' \rangle \sim \frac{1}{N} \tag{34}$$

and contribution of such terms to differential scattering cross-section is $O(\frac{1}{N^2})$ and hence neglected

Case IIIa

$$n_\alpha + n_\beta = n'_\alpha + n'_\beta \quad \text{i.e.} \quad n = n' \pm 1$$

Let $n_{k_1\alpha} = n'_{k_1\alpha}$, $n_{k_1\beta} = n'_{k_1\beta}$ except $n_{k_1',\alpha} = n'_{k_1',\alpha} + 1$

$$\langle A|P^{\alpha'}_{a_J}|A'\rangle$$

$$= \frac{1}{\sqrt{2NJ}}\langle 0|\left[J^{+}_{a_J}, P^{\alpha'}_{J}\right]|0\rangle \sqrt{n_{k_1',\alpha}}\, U_{k_1'}\, e^{i\underline{k}_1' \cdot \underline{R}_J} + O\left(\frac{1}{J^{3/2}}\right) \tag{35}$$

Case IIIb

If $n_{k_1\alpha} = n'_{k_1\alpha}$ and $n_{k_1\beta} = n'_{k_1'}$ e cept

$n_{k_1,\beta} = n'_{k_1,\beta+1}$, then

$$\langle \Lambda | P^{\alpha'}_{a_J} | \Lambda' \rangle$$

$$= \frac{1}{\sqrt{2NJ}} \langle 0 | [J^{-}_{a_J}, P^{\alpha'}_{J}] | 0 \rangle (-v_{k_1'}) \sqrt{n_{l_1,\ell}} \; e^{-i\underline{k}_1' \cdot \underline{R}_J}$$

$$+ O\left(\frac{1}{N^{3/2}} \right) \qquad (36)$$

Case IVa

$n = n'-1$, $n_{k_1\alpha} = n'_{k_1\alpha}$, $n_{k_1\beta} = n'_{k_1\beta}$ e cept

$n_{k_1,\alpha} = n'_{k_1,\alpha-1}$

$$\langle \Lambda | P^{\alpha'}_{a_J} | \Lambda' \rangle$$

$$= \langle 0 | P^{\alpha'}_{a_J} | 1_{k_1,\alpha} \rangle \sqrt{n_{k_1,\alpha}+1} + O\left(\frac{1}{L^{3/2}} \right)$$

$$= \frac{1}{\sqrt{2NJ}} e^{-i\underline{k}_1' \cdot \underline{R}_J} U_{k_1'} \langle 0 | P^{\alpha'}_{a_J} J^{-}_{\gamma_J} | 0 \rangle \sqrt{n_{k_1,\alpha}+1}$$

Case IVb

If $n_{k_1\alpha} = n'_{k_1\alpha}$ and $n_{k_1\beta} = n'_{k_1'}$ e cept

$n_{k_1,\beta} = n'_{k_1,\beta-1}$.

Then

$$\langle A|P^{\alpha'}_{a_J}|A'\rangle$$

$$= \langle 0|P^{\alpha'}_{a_J}|1_{k_1',\beta}\rangle \sqrt{n_{k_1',\beta}+1} + 0\left(\frac{1}{N^{3/2}}\right)$$

$$= \frac{1}{\sqrt{2NJ}}\left(-v_{k_1'}\right) e^{i\underline{k}_1'\cdot\underline{R}_J} \langle 0|P^{\alpha'}_{a_J} J^{+}_{a_J}|0\rangle \sqrt{n_{k_1',\alpha}+1}$$

$$= 0 + 0\left(\frac{1}{N^{3/2}}\right) \qquad (37)$$

Case V

If $|n-n'| = l \geq 2$ say

then

$$\langle l|P^{\alpha}_{a_J}|l'\rangle \sim \frac{1}{N^{l/2}} \qquad (38)$$

and corresponding term in differential scattering cross section is of the order of $\frac{1}{N^l}$ which is neglected as far as one magnon scattering is concerned

To evaluate $\langle l|P^{\alpha'}_{b_1}|l'\rangle$

i.e., $$\langle 0|\left\{\prod_{l}\frac{(\alpha_{k_l})^{n_{k_l\beta}}}{\sqrt{n_{k_l\alpha'}}}\frac{(\beta_{k_l})^{n_{k_l\beta}}}{\sqrt{n_{k_l\beta'}}}\right\}P^{\alpha'}_{b_1}$$

$$\left\{\prod_{l}\frac{(\alpha^{+}_{k_l})^{n_{k_l\alpha}}}{\sqrt{n_{k_l\alpha'}}}\frac{(\beta^{+}_{k_l})^{n_{k_l\beta}}}{\sqrt{n_{k_l\beta'}}}\right\}|0\rangle \qquad (39)$$

Now

$$\prod_{l} (\alpha_{k_l})^{n_{k_l\alpha}} (\beta_{k_l})^{n_{k_l\beta}} P^{\alpha'}_{b_l}$$

$$= \prod_{l} (\alpha_{k_l})^{n_{k_l\alpha}} \Bigg[P^{\alpha'}_{b_l} \prod_{l} (\gamma_{k_l})^{n_{k_l\beta}} + \frac{1}{\sqrt{2NJ}} \left[J^{-}_{b_l}, P^{\alpha'}_{b_l} \right] \times$$

$$\sum_{l} U_{k_l} n_{k_l\beta} (\beta_{k_l})^{n_{k_l\beta}-1} e^{i\underline{k}_l \underline{R}_l} \prod_{l' \neq l} (\gamma_{k_{l'}})^{n_{k_{l'}\beta}}$$

$$+ \frac{1}{2NJ} \frac{\left[J^{-}_{b_l}, \left[J^{-}_{b_l}, P^{\alpha'}_{b_l} \right] \right]}{2} \Bigg\{ \sum_{l} U_{k_l} n_{k_l\beta} (\gamma_{k_l})^{n_{k_l\beta}-1}$$

$$e^{i\underline{k}_l \underline{R}_l} \sum_{l' \neq l} U_{k_{l'}} n_{k_{l'}\beta} \times (\beta_{k_{l'}})^{n_{k_{l'}\beta}-1} e^{i\underline{k}_{l'} \underline{R}_l}$$

$$\prod_{\substack{l'' \neq l \\ \neq l'}} (\gamma_{k_{l''}})^{n_{k_{l''}\gamma}} + \sum_{l} U^2_{k_l} n_{k_l\gamma} (n_{k_l\gamma}-1) e^{2i\underline{k}_l \underline{R}_l}$$

$$(\gamma_{k_l})^{n_{k_l\gamma}-2} \prod_{l' \neq l} (\gamma_{k_{l'}})^{n_{k_{l'}\beta}} \Bigg] + O\left(\frac{1}{N^{3/2}} \right) \qquad (40)$$

$$= P^{\alpha'}_{b_l} \prod_{l} (\alpha_{k_l})^{n_{k_l\alpha}} (\gamma_{k_l})^{n_{k_l\gamma}} + \frac{1}{\sqrt{2NJ}} \left[J^{+}_{b_l}, P^{\alpha'}_{b_l} \right] \sum_{l} (-\nu_{k_l}) \times$$

$$n_{k_l\alpha} (\alpha_{k_l})^{n_{k_l\alpha}-1} \times e^{i\underline{k}_l \underline{R}_l} \prod_{l' \neq l} (\alpha_{k_{l'}})^{n_{k_{l'}\alpha}} \prod_{l_1} (\gamma_{k_{l_1}})^{n_{k_{l_1}\gamma}}$$

$$+\frac{1}{2NJ}\,\frac{\left[J^{-}_{b_1},\left[J^{+}_{b_1},P^{\alpha'}_{b_1}\right]\right]}{2}\left\{\sum_{l_1} v_{k_{l_1}}\, n_{k_{l_1}\alpha}\,(\alpha_{k_{l_1}})^{n_{k_{l_1}\alpha-1}}\right.$$

$$e^{i\underline{k}_{l_1}\underline{R}_{l_1}}\sum_{l_1'\neq l_1} v_{k_{l_1'}}\, n_{k_{l_1'}\alpha}(\alpha_{k_{l_1'}})^{n_{k_{l_1'}\alpha-1}}\, e^{-i\underline{k}_{l_1'}\underline{R}_{l_1}}\prod_{l_1''\neq l_1'}(\alpha_{k_{l_1''}})^{n_{k_{l_1}\alpha}}$$

$$\left.+\sum_{l_1} v^2_{k_{l_1}}\, x\, n_{k_{l_1}\alpha}(n_{k_{l_1}\alpha-1})\, e^{-2i\underline{k}_{l_1}\underline{R}_{l_1}}(\alpha_{k_{l_1}})^{n_{k_{l_1}\alpha-2}}\prod_{l_1'\neq l_1}(\alpha_{k_{l_1'}})^{n_{k_{l_1'}\alpha}}\right\}$$

$$x\prod_{l_1}(\gamma_{k_{l_1}})^{n_{k_{l_1}\gamma}}+\frac{1}{\sqrt{2NJ}}\left[J^{-}_{b_1},P^{\alpha'}_{b_1}\right]\sum_{l_1} U_{k_{l_1}}\, n_{l_1\gamma}\,(\gamma_{k_{l_1}})^{n_{l_1\gamma-1}}$$

$$e^{i\underline{k}_{l_1}\underline{R}_{l_1}}\, x\prod_{l_1'\neq l_1}(\gamma_{k_{l_1'}})^{n_{k_{l_1'}\gamma}}\prod_{l_1}(\alpha_{k_{l_1}})^{n_{k_{l_1}\alpha}}$$

$$+\frac{1}{2NJ}\left[J^{+}_{b_1},\left[J^{-}_{b_1},P^{\alpha'}_{b_1}\right]\right]\sum_{l_1}(-v_{k_{l_1}})\, x\, n_{k_{l_1}\alpha}\,(\alpha_{k_{l_1}})^{n_{k_{l_1}\alpha-1}}$$

$$e^{-i\underline{k}_{l_1}\underline{R}_{l_1}}\prod_{l_1'\neq l_1}(\alpha_{k_{l_1'}})^{n_{k_{l_1'}\alpha}}\sum_{l_1} U_{k_{l_1}}\, n_{l_1\gamma}\,(\gamma_{k_{l_1}})^{n_{l_1\gamma-1}}$$

$$x\, e^{i\underline{k}_{l_1}\underline{R}_{l_1}}\prod_{l_1'\neq l_1}(\gamma_{k_{l_1'}})^{n_{k_{l_1'}\gamma}}+\frac{1}{2NJ}\,\frac{\left[J^{-}_{b_1},\left[J^{-}_{b_1},P^{\alpha'}_{b_1}\right]\right]}{2}\, x$$

$$\left\{\sum_{l_1} U_{k_{l_1}}\, n_{k_{l_1}\gamma}\,(\gamma_{k_{l_1}})^{n_{k_{l_1}\gamma-1}}\, x\, e^{i\underline{k}_{l_1}\underline{R}_{l_1}}\sum_{l_1'\neq l_1} U_{k_{l_1'}}\, n_{k_{l_1'}\gamma}(\gamma_{k_{l_1'}})^{n_{k_{l_1'}\gamma-1}}\right.$$

$$e^{i\underline{k}_{l'}\cdot\underline{P}_l}\prod_{\substack{l''\neq l\\ \neq l'}}(\beta_{k_{l''}})^{n_{k_{l''}}\beta}+\sum_l U_{k_l}^2\, n_{k_l}\,(n_{k_l}\beta-1)$$

$$e^{2i\underline{k}_l\cdot\underline{R}_l}(\beta_{k_l})^{n_{k_l}\beta-2}\prod_{l'\neq l}(\beta_{k_{l'}})^{n_{l'}\beta}\Bigg\}\times\prod_{l_1}(\alpha_{k_{l_1}})^{n_{k_{l_1}}\alpha}$$

$$+O\left(\frac{1}{N^{3/2}}\right) \qquad (41)$$

(41) can be obtained from (32) on making the following replacements

$$P_{a_J}^{\alpha'} \text{ by } P_{b_l}^{\alpha'},\quad U_{k_l} \text{ by } (-v_{k_l}),\quad R_J \text{ by } (-R_l)$$

$$J_{a_J}^- \text{ by } J_{b_l}^- \text{ and } J_{a_J}^+ \text{ by } J_{b_l}^+ \text{ or vice versa}$$

Hence the results of various cases worked out for $\langle A|P_{a_J}^{\alpha'}|A'\rangle$ immediately yield that for $\langle A|P_{b_l}^{\alpha'}|A'\rangle$

Case I

$$\langle A|P_{b_l}^{\alpha'}|A'\rangle$$

$$=\langle 0|P_{b_l}^{\alpha'}|0\rangle+\frac{1}{\sqrt{2NJ}}\langle 0|\left[J_{b_l}^+\,P_{b_l}^{\alpha'}\right]\sum_l|1_{k_l\alpha}\rangle(-v_{k_l})\,n_{k_l\alpha}\,e^{-i\underline{k}_l\cdot\underline{R}_l}$$

$$+\frac{1}{\sqrt{2NJ}}\langle 0|\left[J_{b_l}^-,\,P_{b_l}^{\alpha'}\right]\sum_l|1_{k_l\beta}\rangle U_{k_l}\,n_{k_l\beta}\,e^{i\underline{k}_l\cdot\underline{R}_l}+O\left(\frac{1}{N^2}\right)$$

$$=\langle 0|P_{b_l}^{\alpha'}|0\rangle+\frac{1}{2NJ}\langle 0|\left[J_{b_l}^-,P_{b_l}^{\alpha'}\right]J_{b_l}^+|0\rangle\sum_l U_{k_l}^2\,n_{k_l\beta}+O\left(\frac{1}{N^2}\right) \qquad (42)$$

Case II

$$\langle A|P^{\gamma'}_{b_1}|\Lambda'\rangle \sim \frac{1}{N} \quad \text{same as (34)}$$

Case IIIa

$$\langle A|P^{\alpha'}_{b_1}|\Lambda'\rangle$$

$$= \frac{1}{\sqrt{2NJ}} \langle 0|\left[J^{+}_{b_1} P^{\alpha'}_{b_1} \right]|0\rangle \sqrt{n_{k_1',\alpha}}\,(-v_{k_1'})\, e^{i\underline{k}_1 \cdot \underline{R}_1} + 0\left(\frac{1}{N^{3/2}}\right) \tag{43}$$

Case IIIb

$$\langle A|P^{\alpha'}_{b_1}|\Lambda'\rangle$$

$$= \frac{1}{\sqrt{2NJ}} \langle 0|\left[J^{-}_{b_1}, P^{\alpha'}_{b_1} \right]|0\rangle \sqrt{n_{k_1',\beta}}\, U_{k_1'}\, e^{i\underline{k}_1' \cdot \underline{R}_1} + 0\left(\frac{1}{N^{3/2}}\right) \tag{44}$$

Case IVa

$$\langle A|P^{\alpha'}_{b_1}|\Lambda'\rangle$$

$$= \langle 0|P^{\alpha'}_{b_1}|1_{k_1',\alpha}\rangle \sqrt{n_{k_1',\alpha}+1} + 0\left(\frac{1}{N^{3/2}}\right)$$

$$= 0 + 0\left(\frac{1}{N^{3/2}}\right)$$

Case IVb

$$\langle A|P^{\alpha'}_{b_1}|\Lambda'\rangle$$

$$= \langle 0|P^{\alpha'}_{b_1}|1_{k_1',\beta}\rangle \sqrt{n_{k_1',\beta}+1} + 0\left(\frac{1}{N^{3/2}}\right)$$

$$= \frac{U_{k_1'}}{\sqrt{2NJ}} e^{-i\underline{k}_1' \cdot \underline{R}_1} \langle 0|P^{\alpha'}_{b_1} J^+_{b_1}|0\rangle \sqrt{n_{k_1',\mu}+1} + O\left(\frac{1}{N^{3/2}}\right) \quad (45)$$

Case V

$$\langle A|P^{\alpha}_{b_1}|A'\rangle \sim \frac{1}{N^{1/2}} \quad \text{same as (38)}$$

Now

$$\frac{d^2}{d\Omega\, dE_{p'}}$$

$$= (r_o \hbar)^2 \frac{p'}{p} \sum_{\alpha,\beta} (\delta_{\alpha\beta} - e_\alpha e_\beta) \frac{1}{2\pi\hbar} \int_{-\infty}^{\infty} dt\, e^{i\frac{t}{\hbar}(E_{p'} - E_p)}$$

$$\Bigg[\sum_{J,J'} e^{-i\underline{q}\cdot(\underline{R}_J - \underline{R}_{J'})} \cdot \langle P^{\alpha}_J(0)\, P^{\beta}_{J'}(t)\rangle + \sum_{J,l} e^{-i\,\underline{q}\cdot(\underline{R}_J - \underline{R}_l)} \langle P^{\alpha}_J(0)\, P^{\beta}_l(t)\rangle + \sum_{l,J} e^{-i\underline{q}\cdot(\underline{R}_l - \underline{R}_J)} \langle P^{\alpha}_l(0)\, P^{\beta}_J(t)\rangle + \sum_{l,l'} e^{-i\underline{q}\cdot(\underline{R}_l - \underline{R}_{l'})} \langle P^{\alpha}_l(0)\, P^{\beta}_{l'}(t)\rangle \Bigg] \quad (46)$$

Now, taking $|0\rangle = \prod_{J,l} |JJ\rangle_{aJ} |J\text{-}J\rangle_{bl}$

we have

Case I

$$\langle A|P^{\alpha'}_{a_J}|A'\rangle$$

$$= \langle JJ|P^{\alpha'}_{a_J}|JJ\rangle_J + \frac{1}{N}\sum_1 U^2_{k_1}\, n_{k_1\alpha}\left\{ {}_J\langle JJ-1|P^{\alpha'}_{a_J}|JJ-1\rangle_J - {}_J\langle JJ|P^{\alpha'}_{a_J}|JJ\rangle_J\right\} \tag{47}$$

and

$$\langle A|P^{\alpha'}_{b_1}|A'\rangle = {}_1\langle J-J|P^{\alpha'}_{b_1}|J-J\rangle_1 + \frac{1}{N}\sum_{\ell} U^2_{k_1}\, n_{k_1\beta}\left\{ {}_1\langle J,-J+1|P^{\alpha'}_{b_1}|J-J+1\rangle_1 - {}_1\langle J-J|P^{\alpha'}_{b_1}|J-J\rangle_1\right\} \tag{48}$$

Case IIIa

$$\langle A|P^{\alpha'}_{a_J}|A'\rangle = \frac{1}{\sqrt{N}}\, {}_J\langle JJ-1|P^{\alpha'}_{a_J}|JJ\rangle_J \sqrt{n_{k_1',\alpha}}\, U_{k_1'}\, e^{i\underline{k}'_1\cdot\underline{R}_J}$$

$$\langle A|P^{\alpha'}_{b_1}|A'\rangle = \frac{1}{\sqrt{N}}\, {}_1\langle J,-J|P^{\alpha'}_{b_1}|J,-J+1\rangle_1 \sqrt{n_{k_1',\alpha}}\, v_{k_1'}\, e^{-i\underline{k}_1'\cdot\underline{R}_1} \tag{49}$$

Case IIIb

$$\langle A|P^{\alpha'}_{a_J}|A'\rangle = \frac{1}{\sqrt{N}}\, {}_J\langle JJ|P^{\alpha'}_{a_J}|JJ-1\rangle_J\, v_{k_1'} \sqrt{n_{k_1',\beta}}\, e^{-i\underline{k}_1'\cdot\underline{R}_J}$$

$$\langle A|P^{\alpha'}_{b_1}|A'\rangle = \frac{1}{\sqrt{N}}\, {}_1\langle J,-J+1|P^{\alpha'}_{b_1}|J-J\rangle_1 \sqrt{n_{k_1',\beta}}\, U_{k_1'}\, e^{i\underline{k}_1'\cdot\underline{R}_1} \tag{50}$$

Case IVa

$$\langle A|P^{\alpha'}_{a_J}|A'\rangle = \frac{1}{\sqrt{N}} e^{-i\underline{k}_{l'}\cdot\underline{R}_J} U_{k_{l'}} {}_J\langle JJ|P^{\alpha'}_{a_J}|J,J-1\rangle \sqrt{n_{l_{l'},\alpha\mp 1}}$$

$$\langle A|P^{\alpha'}_{b_l}|A'\rangle = 0 \qquad (51)$$

Case IVb

$$\langle A|P^{\alpha'}_{a_J}|A'\rangle = 0 \qquad (52)$$

$$\langle A|P^{\alpha'}_{b_l}|A'\rangle = \frac{1}{\sqrt{N}} {}_l\langle J,-J|P^{\alpha'}_{b_l}|J,-J+1\rangle_l U_{k_{l'}} \sqrt{n_{k_{l'},\beta+1}}\, e^{-i\underline{k}_{l'}\cdot\underline{R}_l}$$

Now

$$\langle P^{\alpha'}_{a_J} P^{\beta'}_{b_l}(t)\rangle$$

$$= \sum_{\Lambda,\Lambda'} e^{-E_{\Lambda\beta'}} \langle\Lambda|P^{\alpha'}_{a_J}|\Lambda'\rangle\langle\Lambda'|P^{\beta'}_{b_l}|\Lambda\rangle \exp\frac{\left[\frac{1}{\hbar}\int_0^t (E,-\omega)\right]}{\sum e^{-E_{\Lambda\beta'}}}$$

$$= {}_J\langle JJ|P^{\alpha'}_{a_J}|JJ\rangle_J\, {}_l\langle J,-J|P^{\beta'}_{a_J}|J-J\rangle_l + \sum_l \frac{U^2_{b_l}}{N}\langle n_{l_l\alpha}\rangle$$

$$\times {}_l\langle J-J|P^{\beta'}_{b_l}|J-J\rangle_l \left\{ {}_J\langle JJ-1|P^{\alpha'}_{a_J}|JJ-1\rangle_J - {}_J\langle JJ|P^{\alpha'}_{a_J}|JJ\rangle_J \right\}$$

$$+ \frac{1}{N}\sum_l U^2_{k_l}\langle n_{k_l\beta}\rangle \times {}_J\langle JJ|P^{\alpha'}_{a_J}|JJ\rangle_J \left\{ {}_l\langle J,-J+1|P^{\beta'}_{b_l}|J,-J+1\rangle_l - {}_l\langle J-J|P^{\beta'}_{b_l}|J-J\rangle_l \right\} + \frac{1}{N} {}_J\langle JJ|P^{\alpha'}_{a_J}|JJ-1\rangle_J\, {}_l\langle J,-J|P^{\beta'}_{b_l}|J,-J+1\rangle_l \times$$

$$\sum_{l} v_{k_l'} U_{k_l'} \langle n_{k_l',\beta} \rangle e^{-i\underline{k}_l'(\underline{R}_J+\underline{R}_l)} \times e^{\frac{i}{\hbar}t(-\epsilon_{k_l',\beta})} + \frac{1}{iJ} \times$$

$$J \langle JJ|P^{\alpha'}_{a_J}|J,J-1\rangle_J \; l \langle J,-J|P^{\beta'}_{b_l}|J,-J+1\rangle_l \sum_{l'} U_{l\,l'} v_{l\,l'} (\langle n_{l\,l',\alpha}\rangle+1)$$

$$\times e^{-i\underline{k}_l'(\underline{R}_J+\underline{R}_l)} e^{\frac{i}{\hbar}t\epsilon_{k_l',\alpha}} \qquad (53)$$

First two terms of $\langle P^{\alpha'}_{b_l} P^{\beta'}_{a_J}(t)\rangle$ are obtained from above by simply interchanging α' and β'

$$\langle P^{\alpha'}_{a_J} P^{\beta'}_{a_J'}(t)\rangle$$

$$= J\langle JJ|P^{\alpha'}_{a_J}|JJ\rangle_J \; J'\langle JJ|P^{\beta'}_{a_J'}|JJ\rangle_{J'} + \sum_{l} \frac{U^2_{k_l}\langle n_{k_l\alpha}\rangle}{N} \Big\{ J\langle JJ|P^{\alpha'}_{a_J}|JJ\rangle_J$$

$$\times \left(J'\langle JJ-1|P^{\beta'}_{a_J'}|JJ-1\rangle_{J'} - J'\langle JJ|P^{\beta'}_{a_J'}|JJ\rangle_{J'}\right) + J'\langle JJ|P^{\beta}_{a_J'}|JJ\rangle_J$$

$$\times \left(J\langle JJ-1|PP^{\alpha}_{a_J}|JJ-1\rangle_J - J\langle JJ|P^{\alpha}_{a_J}|JJ\rangle_J\right)\Big\} + \frac{1}{N} J\langle JJ-1|P^{\alpha'}_{a_J}|JJ\rangle_J$$

$$\times J'\langle JJ|P^{\beta'}_{a_J'}|JJ-1\rangle_{J'} \sum_{l'} \langle n_{k_l',\alpha}\rangle U^2_{k_l'} e^{i\underline{k}_l'(\underline{R}_J-\underline{R}_{J'})+\frac{i}{\hbar}t(-\epsilon_{k_l',\alpha})}$$

$$+ \frac{1}{N} J\langle JJ|P^{\alpha'}_{a_J}|JJ-1\rangle_J \times J'\langle JJ-1|P^{\beta'}_{a_J'}|JJ\rangle_{J'} \sum_{l'} (\langle n_{l\,l',\alpha}\rangle+1)$$

$$U^2_{k_l'} e^{-i\underline{k}_l'(\underline{R}_J-\underline{R}_{J'})} e^{\frac{i}{\hbar}t\epsilon_{k_l',\alpha}} \qquad (54)$$

$$< P^{\alpha'}_{b_1} P^{\beta'}_{b_{1'}}(t)$$

$$= {}_1\langle J-J|P^{\alpha'}_{b_1}|J-J\rangle_1 \; {}_{1'}\langle J-J|P^{\beta'}_{b_{1'}}|J-J\rangle_{1'} + \frac{1}{N}\sum_{1} U^2_{k_1} \langle n_{k_1\beta}\rangle$$

$$\Big[{}_1\langle J-J|P^{\alpha'}_{b_1}|J-J\rangle_1 \Big\{ {}_{1'}\langle J,-J+1|P^{\beta'}_{b_{1'}}|J,-J+1\rangle_{1'} - {}_{1'}\langle J,-J|P^{\beta'}_{b_{1'}}|J,-J\rangle_{1'} \Big\}$$

$$+ {}_{1'}\langle J-J|P^{\beta'}_{b_{1'}}|J-J\rangle_{1'} \Big\{ {}_1\langle J,-J+1|P^{\alpha}_{b_1}|J,-J+1\rangle_1 - {}_1\langle J,-J|P^{\alpha}_{b_1}|J,-J\rangle_1 \Big\} \Big]$$

$$+ \frac{1}{N} {}_1\langle J,-J+1|P^{\alpha'}_{b_1}|J,-J\rangle_1 \; {}_{1'}\langle J,-J|P^{\beta'}_{b_{1'}}|J,-J+1\rangle \times \sum_{1'} U^2_{k_{1'}}$$

$$\langle n_{k_{1'}\beta}\rangle \; e^{i\underline{k}_{1'}(\underline{R}_1-\underline{R}_{1'})} \; e^{\frac{i}{\hbar}t(-\epsilon_{k_{1'}\beta})} + \frac{1}{N} {}_1\langle J-J|P^{\alpha'}_{b_1}|J,-J+1\rangle_1$$

$$\times \; {}_{1'}\langle J,-J+1|P^{\beta'}_{b_{1'}}|J-J\rangle_{1'} \sum_{1'} (\langle n_{k_{1'}\beta}\rangle + 1) U^2_{k_{1'}}$$

$$e^{-i\underline{k}_{1'}(\underline{R}_1-\underline{R}_{1'})} \; e^{\frac{i}{\hbar}t\epsilon_{k_{1'}\beta}} \qquad (55)$$

It is to be noted that on performing the summation over R_1 and R_J the third and fourth term of (53) will vanish due to occurrence of $e^{ik_1(R_1 + R_J)} \times e^{-iq(R_1-R_J)}$

4 3 ONE MAGNON INTENSITIES

Each magnetic unit cell has two ions, one at $\bar{R}_J$, other at the $\bar{R}_1 = \bar{R}_J + \bar{r}_{12}$ So

$$\left(\frac{d^2\sigma}{d\Omega dE_{p'}}\right)_{elec}$$

$$= (r_o\gamma)^2 \sum_{\alpha\beta} (\delta_{\alpha\beta} - e_\alpha e_\beta)\, S(E_{p'}, -\Box_p) \quad \frac{(2\pi)^3 N_m}{v_o m} \sum_{\underline{\tau}} \delta(\underline{q} - \underline{\tau})$$

$$\times \Big[\Big\{ {}_J\langle JJ|P^\alpha_{a_J}|JJ\rangle_J \; {}_{J'}\langle JJ|P^\beta_{a_{J'}}|JJ\rangle_{J'} + {}_1\langle J,-J|P^\alpha_{b_1}|J,-J\rangle_1$$

$${}_{1'}\langle J,J|P^\beta_{b_{1'}}|J,-J\rangle_{1'} \Big\} \times \Big\{ 1 - \frac{2}{N}\sum_1 U^2_{k_1} \langle n_{k_1}\rangle \Big\}$$

$$+ 2\sum_1 \frac{U^2_{k_1}\langle n_{k_1}\rangle}{N} \Big\{ {}_J\langle JJ|P^\alpha_{a_J}|JJ\rangle_J \; {}_{J'}\langle JJ-1|P^\beta_{a_{J'}}|JJ-1\rangle_{J'}$$

$$+ {}_1\langle J-J|P^\alpha_{b_1}|J-J\rangle_1 \; {}_{1'}\langle J,-J+1|P^\beta_{b_{1'}}|J,-J+1\rangle_{1'} \Big\}$$

$$+ 2\cos \underline{\varrho}\, \underline{r}_{12} \Big\{ {}_J\langle JJ|P^\alpha_{a_J}|JJ\rangle_J \times {}_1\langle J,-J|P^\beta_{a_1}|J,-J\rangle_1$$

$$\Big(1 - \frac{2}{N}\sum_1 U^2_{k_i}\langle n_{k_1}\rangle\Big) + \frac{1}{N}\sum_1 U^2_{k_1}\langle n_{k_1}\rangle \, ({}_1\langle J-J|P^\beta_1|J-J\rangle_1$$

$$\times {}_J\langle JJ-1|P^\alpha_{a_J}|JJ-1\rangle_J + {}_J\langle JJ|P^\alpha_{a_J}|JJ\rangle_J \; {}_1\langle J,-J+1|P^\beta_{b_1}|J,-J+1\rangle_1) \Big\} \Big]$$

Because $\langle n_{k_1\alpha}\rangle = \langle n_{k_1\beta}\rangle = \dfrac{1}{\epsilon_{k_1\beta} - 1}$ as $k_1\alpha = k_1\beta$ for antiferromagnet,

$$\left(\frac{d^2\sigma}{d\Omega dE_{p'}} \right)_{\pm 1}$$

$$= (r_o\gamma)^2 \sum_{\alpha\beta} (\delta_{\alpha\beta} - e_\alpha e_\beta) \frac{(2\pi)^3}{V_{om}} \left[\sum_{\underline{z},k_1} \delta(\underline{q} - \underline{k}_1 - \underline{z})\, \delta(E_{p'} - E_p - \epsilon_{k_1}) \right.$$

$$\times \langle n_{k_1} \rangle U_{k_1}^2 \left\{ {}_J\langle JJ-1 | P^\alpha_{a_J} | JJ \rangle_J \; {}_{J'}\langle JJ | P^\beta_{a_{J'}} | JJ-1 \rangle_{J'} \right.$$

$$\left. + {}_1\langle J,-J+1 | P^\alpha_{b_1} | J,-J \rangle_1 \times {}_{1'}\langle J,-J | P^{\beta'}_{b_{1'}} | J,-J+1 \rangle_{1'} \right\}$$

$$+ \sum_{\underline{z}} \sum_{k_1} \delta(\underline{q} + \underline{k}_i - z)\, \delta(E_{p'} - E_p + \epsilon_{k_1}) \times \left\{ {}_J\langle JJ | P^\alpha_{a_J} | JJ-1 \rangle_J \right.$$

$$\left. {}_{J'}\langle JJ-1 | P^\beta_{a_{J'}} | JJ \rangle_{J'} + {}_1\langle J,-J | P^\alpha_{b_1} | J,-J+1 \rangle_1 \times {}_{1'}\langle J,-J+1 | P^{\beta'}_{b_{1'}} | J,-J \rangle_{1'} \right\}$$

$$(\langle n_{k_1} \rangle + 1)\, U^2_{k_1}$$

$$= (r_o\gamma)^2 \sum_{\alpha\beta} (\delta_{\alpha\beta} - e_\alpha e_\beta) \frac{(2\pi)^3}{V_{om}} \left\{ {}_J\langle JJ-1 | P^\alpha_{a_J} | JJ \rangle_J \right.$$

$$ {}_{J'}\langle JJ | P^\beta_{a_{J'}} | JJ-1 \rangle_{J'} + {}_1\langle J,-J+1 | P^\alpha_{b_1} | J-J \rangle_1$$

$$\left. {}_{1'}\langle J,-J | P^{\beta'}_{b_{1'}} | J,-J+1 \rangle_{1'} \right\} \sum_1 U_1^2 \sum_{\underline{z}} \left\{ \langle n_{k_1} \rangle \delta(\underline{q} - \underline{k}_1 - \underline{z}) \right.$$

$$\left. \delta(E_{p'} - E_p - \epsilon_{k_1}) + (\langle n_{k_1} \rangle + 1)\, \delta(\underline{q} + \underline{k}_1 - \underline{z})\, \delta(E_{p'} - E_p + \epsilon_{k_1}) \right\}$$

$$N = Nm$$

CHAPTER - V

CONCLUSION

The simple ordered Neel state is not the ground state of a real antiferromagnetic system The correct ground state is

$$|O\rangle = C \exp \left[\sum_{p,q,k} M_k^{pq} a_r^{(p)} a_s^{(q)*} \right] |G\rangle$$

where $|G\rangle$ is the ordered state, $a_k^{(p)}$ is identical with C_k of equation (19) of the last chapter (p) and (q) represent generalization to many sublattice system For a two sublattice system $p = 1,2$ and $q = 1,2$ $M_{rs} = (\underline{\underline{\alpha}})^{-1} (\underline{\underline{\beta}})$ where $\underline{\underline{\alpha}}$ and $\underline{\underline{\beta}}$ are transformation matrices utilized in diagonalizing the Hamiltonian (L R Walker, Spin Waves and other magnetic modes, Magnetism, Vol I, Ed G T Rado and H Suhl, Academic Press, N Y 1963, p 315) For using in the analysis of experimental data, the theory of the scattering of neutrons from antiferromagnetic substances developed in the last chapter has to be modified by replacing the ordered ground state by the correct ground state described above The work, along with its application to real antiferromagnetics with negligible anisotrophy (e g , $KCoF_3$), is in progress

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